The Effects of Long Range Transport of Air Pollutants on Artic Cloud-Active Aerosol

by
Randolph D. Borys

Department of Atmospheric Science
Colorado State University
Fort Collins, Colorado

Colorado State University

Department of Atmospheric Science

Paper No. 367

THE EFFECTS OF LONG-RANGE TRANSPORT OF AIR POLLUTANTS ON ARCTIC CLOUD-ACTIVE AEROSOL

By

Randolph Dean Borys

This report was prepared with support provided by Graduate School of Oceanography, University of Rhode Island, Office of Naval Research Contract N0014-76-C-0435, and Department of Atmospheric Science, Colorado State University, National Science Foundation Grants ATM 7819261 and ATM 8109490, Principal Investigator, Lewis O. Grant

Department of Atmospheric Science
Colorado State University
Fort Collins, Colorado

May 1983

Atmospheric Science Paper No. 367

ABSTRACT OF THESIS

THE EFFECTS OF LONG-RANGE TRANSPORT
OF AIR POLLUTANTS ON ARCTIC CLOUD-ACTIVE AEROSOL

Arctic Haze, observed over much of the Arctic during winter, consists of submicron primary and secondary pollutant aerosol particles transported from mid-latitude source regions. These pollutants, if present would alter the concentration of cloud-active aerosols. Cloud-active aerosol particles are responsible for the heterogeneous nucleation of water (cloud condensation nuclei, CCN) and ice (ice nuclei, IN) in the atmosphere and are responsible for the initial microphysical structure of clouds. Cloud microstructure determines, in part, the radiative properties and colloidal stability of a cloud. A change in the climatically important cloud-active aerosol particle concentration over an extended period would alter the radiation balance, leading to a change in temperature. This would change the pole to equator temperature gradient which is the driving force of the atmospheric general circulation.

A series of studies was conducted at several arctic locations to determine the relationship between CCN and IN and the pollutant aerosol. The trace elemental composition of the total aerosol was used to determine when anthropogenic aerosol was present. Variations of the elemental composition of the aerosol and concentrations of the cloud-active aerosol particles were compared. The arctic sites were chosen to lie along proposed pollutant aerosol transport paths to the Arctic.

Various comparisons between pollutants and cloud active aerosols have been made on a day by day basis and for summer and winter seasons. The differences between the nucleational properties of Arctic Haze aerosol, which is pollution derived, and the natural background aerosol of the Arctic have been determined.. The elemental composition of the aerosols, combined with meteorological analysis, have been used to assess the relative importance of the pathways.

Results show the CCN concentrations are positively correlated with the pollutant aerosol concentration on both a daily and seasonal basis. Concentrations of IN were found to be lowest during the winter when pollutant aerosol was present. Day-to-day variations of IN were related to both natural and pollutant aerosol sources depending upon the location and season. The lower IN concentrations which occurred during winter when pollutant aerosol concentrations were at a maximum may be a result of preferential removal of IN in the cold Arctic atmosphere or a 'poisoning' of the nuclei by the pollutants. Interpretation of the results provides evidence that pollutant aerosol transport from Eurasia directly to the north is primarily responsible for Arctic Haze.

The effects of pollutant transport to the Arctic on cloud microstructure may be significant. If the results of the surface-based cloud-active aerosol measurements can be extrapolated to arctic clouds, and if homogeneous ice nucleation is not a major determinant for Arctic cloud ice crystal concetrations, clouds which form in the polluted arctic airmass might have fewer ice crystals and more cloud drops per unit volume of cloud. The resulting clouds would be more colloidally

stable and optically thick than clouds which would have formed had no pollutants been present.

Randolph Dean Borys
Department of Atmospheric Science
Colorado State University
Fort Collins, CO 80523
Spring, 1983

ACKNOWLEDGEMENTS

I would like to acknowledge the helpful suggestions and support of my graduate committee; Drs. Elmar Reiter and Rod Skogerboe for their insight in the areas of general circulation, atmospheric chemistry and statistics, Dr. Ken Rahn without whose inspiration and continued support the research would not have been possible, and Professor Lew Grant, who gave me the freedom to pursue my research goals.

Special thanks go to Ruta Radziunas for her long hours on the word processor typing the first draft, and for the energy she seemed to give to those around her. Thanks to Brenda Thompson, the final draft was completed by the deadline. Jan Davis did the finishing touches while I was away on more field work and thanks to to her for getting my thesis to the Graduate School. I would also like to thank Lucy McCall for the excellent drafting work which she did.

I acknowledge and thank all the friends and acquaintances who I met during the course of the work and who gave me support, encouragement and inspiration and helped toward the completion of what often seems to be a never-ending task.

Finally, I will always be indebted to my wife and partner in life, Marilyn, who put up with all the trials and tribulations of being a 'field-widow' for far too long. Her continued support and caring were well beyond what anyone could expect and will serve as an example for me in the future.

TABLE OF CONTENTS

I. INTRODUCTION

The Arctic classically conjures up an image of a vast pristine region, untouched by activities of man. This usually includes an atmosphere of unusual clarity, permitting unrestricted visibility even beyond the horizon due to the refraction of light. This conception has largely been the result of reports of early explorers and visitors to this day who had typically visited the Arctic during the summer-half of the year. The first observations which contradicted this picture were made during weather reconnaissance flights conducted by the U. S. Air Force during the 1950's over the pack-ice north of Alaska (Mitchell, 1956). In some instances, slant-path visibilities were restricted to 3 to 8 km. As a result of the frequent observation of reduced visibility, the phenomenon became known as "Arctic Haze". It is defined in the Glossary of Meteorology published by the American Meteorological Society as a condition of reduced horizontal and slant visibility encountered by aircraft over arctic regions. Visibility in the verticle is unimpeded. It has no distinct upper or lower boundaries and appears greyish-blue when viewed away from the sun and reddish-brown when viewed into the sun. No optical phenomena are produced that would be expected if the haze were composed of ice crystals.

Later, measurements of turbidity made in Canada and northern Alaska at the surface showed unexplained high values for such a remote region (Holmgren, 1971; Shaw and Wendler, 1972; and Shaw, 1975). Subsequently, aerosol measurements made at Barrow, Alaska have shown that man-made

aerosols are an important component of the total aerosol during the late fall, winter and early spring (Rahn et al., 1977). These observations have been confirmed by investigators from Canada and Scandinavia as being typical of their regions in the Arctic (Barrie et al., 1981; Heidam, 1981 and Ottar, 1981).

Whereas up to now studies in the Arctic have concentrated on the chemistry of the aerosol, this study focuses on the nucleational properties of the aerosol. The intensity, persistence and widespread distribution of the haze and its parent aerosol will cause perturbations on the arctic radiation balance. The visible manifestation of the aerosol in the form of haze is primarily due to the aerosol interacting with water vapor from the atmosphere and nucleating liquid water droplets and ice crystals. From a climatic standpoint, it is important to investigate in more detail the nature of this interaction between arctic aerosol and water vapor. Cloud droplet and ice crystal concentrations and size distributions determine the reflectivity and transmissivity of clouds. Clouds are a major factor in determining the global albedo. This study investigates the fraction of the aerosol responsible for the formation and microstructure of arctic clouds. These are the cloud-active aerosol, which are defined as the dry particles which, when in the presence of a sufficient water-vapor density in the atmosphere, will initiate the growth of liquid water droplets and/or ice crystals. By definition, these are cloud-condensation nuclei (CCN) and ice nuclei (IN). The importance of these nucleants cannot be underestimated. They are responsible for the basic cloud microstructure, which determines the interaction of the cloud with solar and terrestrial radiation. Increases in cloud particle

concentrations (water and ice) while increasing the optical thickness of the cloud, will also increase its reflectivity and lower its absorptivity. Thesis effect occur for radiation with wavelength in the near infrared including portions of both the solar and terrestrial radiation spectra. Cloud microstructure also deterines the colloidal stability of the cloud which in part determines the lifetime of the cloud.

An understanding of the relationship between the observed anthropogenic aerosol and the cloud-active aerosol can serve as a basis for estimating the influence of man on the spatial and temporal variations of cloud microstructure. The Arctic provides a unique opportunity for the study of the influences of man-made pollutants in the atmosphere. It is remote from pollutant sources and yet it is influenced by pollutants which have been advected into the Arctic over long distances. The Arctic itself has very few sources of man-made pollutants. Much of the year the Arctic is covered with a layer of snow and ice which serves to inhibit natural aerosol particle formation from biogenic, oceanic or continental sources. The situation is much like a large atmospheric mixing bowl where aerosol is advected to the region rather than generated within. The Arctic is also a desert. Natural removal of aerosol by precipitation is not as rapid as in more southern latitudes. The Arctic atmosphere is also relatively thermally stable in its vertical structure. Vertical mixing of the atmosphere is not strong. Layers of haze are typically observed in the Arctic atmosphere. All of these characteristics provide a relatively simple atmosphere within which to conduct aerosol experiments. The strong influence of man-made pollutants in the Arctic atmosphere and the strong seasonal

dependence of these pollutants as shown by other investigators, provides a natural laboratory situation for the study of the atmospheric effects of the pollutants on the region. Since the Arctic is a large region, and the pollutants are apparently widely dispersed, study of the effects of the pollutants would have application over an extensive area of the globe.

This study was designed to take advantage of the favorable conditions for the study of air pollutants in the Arctic on the concentrations of cloud condensation nuclei and ice nuclei. The surface-based measurements made at five Arctic locations were brought together to describe what role the pollutants play in determining the total Arctic cloud-active aerosol concentrations. The coherence of the results from location to location show that they are applicable over the entire Arctic region. The advection of pollutants to the Arctic is linked to an increase in cloud condensation nuclei concentrations. Conversely, ice nuclei concentrations are lower when the pollutants are present. Since the pollutants are present primarily during the winter, one component of the lowered ice-nuclei concentrations may result from selective removal due to the colder atmospheric conditions. The lowered ice nuclei concentrations can be linked to the pollutants independent of temperature.

This study showed that in remote regions an experiment can be designed combining both chemical and physical measurements of the aerosol to derive a more complete understanding of the properties of the aerosol and the role which man plays in determining these characteristics.

II. HISTORICAL BACKGROUND

2.1 Chemistry of the Arctic Aerosol

Actual direct measurement of the chemical composition of the arctic aerosol did not begin until quite recently. Even after the arctic haze observations made by the U. S. Air Force during weather reconnaissance flights over the Arctic Ocean, there were no investigations directed toward determining the origins of the haze. At about the same time, interest in the aerosol in remote background regions began and various studies were conducted on the Greenland icecap in conjunction with the overall goals of the IGY (1957-8).

During the summers of 1957-8, Fenn and Weickmann (1959) measured the concentration of aerosol particles near Thule, Greenland. They found that their instrumentation was not sensitive enough to give reliable values for the number concentration of the aerosol particles. These investigations were followed by those of Fenn (1960) using more sensitive instrumentation which allowed the determination of the aerosol size distribution. These measurements were made at the Fist Clench site, 210 miles east of Thule, during summer and at Camp Century, 150 miles east of Thule, during winter. They were unable to detect any aerosol particles during the winter period and found a very stable aerosol particle concentration during the summer. Fenn speculated on the origin of these aerosol particles, citing the results of Junge (1960), who determined the chemical composition of ice cores from a site 200 miles east of Thule.

The study by Junge (1960) was the first systematic search for an effect of man's activities on the chemical composition of the atmosphere (aerosol) in the Arctic. Assuming the composition of the ice would reflect the composition of the aerosol at the time the ice was deposited, the ice was analyzed for $SO_4^=$ and Na, indicators of a combination of natural-marine sources and secondary-anthropogenic aerosol (see Chapter I). The ice core permitted analyses to be dated back to 1915. Results showed no increase in $SO_4^=$ due to man, even though he estimated there should have been an increase of 25 per cent based upon fossil fuel consumption increases since that time. He also noted that the $SO_4^=$ concentration was ten times that of any other constituent measured (Cl^-, Na^+, Ca^+, K^+).

This prompted Fenn et al. (1963) to measure the aerosol composition directly. Results from a summer field experiment at Camp Century showed the $SO_4^=$ concentration to average 0.4 $\mu g\ m^{-3}$, which they estimated to comprise 40 per cent of the total aerosol mass. This agreed roughly with Junge's estimates. During the month of January, 1965, Winchester and Duce (1966) and Duce et al. (1966) reported on the iodine, bromine and chlorine content of aerosols and snow collected at Barrow and Fairbanks, Alaska. Their samples for aerosols, collected at the surface and aloft at a few hundred meters altitude, indicated that iodine and bromine were associated with the submicron size range whereas the chlorine was associated with larger aerosol. They did not report on any other constituents of the aerosol and speculated that a vapor-phase distillation process was responsible for the presence of iodine and bromine on the smaller particles. They ruled out the possible motor-fuel source of bromine. They observed similar iodine and bromine

concentrations in snow at Fairbanks, Alaska as in Hawaii, although the concentrations of chlorine were much lower. This supported their aerosol studies in that the larger size chlorine-containing aerosol would have been removed more quickly than the smaller-sized iodine and bromine-containing aerosol, assuming a distant marine source. Snow analyses at Barrow showed that the halogen content was dominated by the proximity of the site to the coast of the Chukchi Sea, a source of sea-salt aerosol. These results suggested that the halogen composition of the Barrow aerosol may be dominated by local sea-salt aerosol production. However, iodine and bromine had the potential of a long-range-transport component.

Murozumi et al. (1969) also studied ice cores collected near Camp Century and found lead concentrations in the ice today to be well over 500 times that found in ice deposited about 800 BC. Later, these same samples and those of Hansen and Langway (1966) were analyzed by Koide and Goldberg (1971) for Cl^- and $SO_4^=$. Subtracting out the sea-salt component of the $SO_4^=$, they found the lead from the same samples to co-vary with the residual $SO_4^=$ for the most recent snow samples (1964-5). This residual or excess $SO_4^=$ comprised 60 per cent of the total $SO_4^=$. They attributed this excess $SO_4^=$ to anthropogenic sources. They found the lead and $SO_4^=$ to have higher concentrations in the winter than in the summer. They speculated that the source of these materials could be the U.S., and ascribed the seasonal dependence to higher wind speeds during the winter, which allowed longer transport distances.

Further work by Herron et al. (1977) on ice cores from Station Milcent in central Greenland also showed the increased effects of man. Zinc, lead and sulphur were found to be present in higher concentrations

in current snow than in snow dated to pre-1900. Cadmium did not show this trend. Vandium was found in proportions explainable by natural dust sources. The authors explained this by the relative volatility of Zn, Pb and S in comparison to V, and that, apparently, natural sources of Cd overshadowed man-made emissions. Their overall conclusion was that for some elements, the effects of man's activities could be detected, but that natural high-temperature processes, such as volcanism, could explain the occurence of other elements "enriched" relative to natural dusts.

Aerosol particle composition in the Soviet Arctic was studied by Egorov et al. (1970). They reported the aerosol trace-element composition for various sites in the U.S.S.R. including three sites at or north of the Arctic Circle: the city of Salekhard, Dickson Island and Novaya Zemlya. Their results showed that the Arctic site on Novaya Zemyla had the lowest trace-element concentrations of all the locations studied and little if any seasonal variability. Further analysis of the data of Egrov et al. by Rahn (1976) shows that a different picture emerges if one looks at the enrichment factor (EF) of the elements Cr, Mn, Ni, Cd and Sn. For the northernmost site of Novaya Zemyla (72^{o} N. Lat.) the mean annual enrichment factors of these elements rank among the ten highest ever measured, including urban areas. High enrichment factors of certain elements can be used as an indication of the presence of pollution aerosol particles. These results must be interpreted cautiously because of local mining and smelting industries in the area. The fact that such heavy industry does exist so far north is important in the interpretation of later aerosol measurements.

Continued interest in the Greenland area as a background location for aerosol measurements prompted Danish investigators to conduct two aircraft experiments over the Greenland icecap in the summers of 1971 and 1973 (Flyger et al., 1973 and Flyger et al., 1976). Results from these flights were inconclusive concerning the aerosol composition. Samples were too small for reliable chemical analysis. In general, they found low overall concentrations of aerosol in all size ranges. Particulate $SO_4^{=}$, and SO_2 gas, were found to increase in concentration with height, which suggested transport to Greenland aloft. Haze layers were observed aloft on the second expedition at altitudes of 8,000 to 10,000 feet. These corresponded to elevated concentrations of Aitken and large nuclei. Air-mass trajectories indicated that the air originated in the Hudson Bay area which suggested that the haze was not of a direct anthropogenic origin. Without further compositional analysis, no conclusions as to the the source of the haze could be drawn.

Measurements were also conducted on the ground during the 1973 experiment. Comparisons of Aitken nuclei concentrations measured at the surface and at the top of a 20-m tower showed consistently higher values on the tower. This observation implies the existence of a shallow surface boundary layer and suggests that measurements made close to the surface may not represent the atmosphere a short distance above. This is certainly true where long downslope trajectories of the air are possible, such as katabatic winds on an icecap, which warm the air by compression and forms a temperature gradient so that the snow surface acts as a thermal precipitator.

As a result of the difficulties with airborne aerosol sampling, Flyger and Heidam (1978) set up a ground station in North Greenland at Kap Harald Moltke for two months during the summer of 1974. Results of the sampling, which extended over several days, showed the elements Zn, Pb, Br, S, Cl and Se to be enriched relative to a crustal-dust source. They point out that the high EF's of the first three of these elements may be due to natural volcanic emissions as suggested by Duce et al. (1975) and Mroz and Zoller (1975). Based upon the results of Koide and Goldberg (1971), however, they believed the anthropogenic component of the S measured was as important as natural sources since it was the second most abundant element in the aerosol. They noted that the concentration of S was only one-third of that found in airborne measurements over Greenland (Flyger et al., 1976).

All of the investigations up to this point were independent attempts at determining the "background" aerosol chemistry of the Arctic. All of the sucessful direct measurements of the aerosol were confined to the summer season and continued for only a few months at most. The few attempts during the winter apparently failed due to instrumental inadequacies. None of the results directly addressed the question concerning the source of Arctic Haze. The topic of Arctic Haze had not been discussed in the literature since it was introduced by Mitchell (1956). Renewed interest was sparked by measurements of high turbidities made in the Alaskan Arctic by Shaw and Wendler (1972) and Shaw (1975). Late-spring values for turbidity were of the order of those found at mid-latitude regions. Vertical profiles of the Mie extinction coefficient were determined by Holmgren et al. (1974). These

profiles indicated a fairly broad haze layer aloft. These investigators also noted that turbidities decreased during the summer season.

The first study directed specifically toward determining the cause of Arctic haze was conducted in 1976 (Rahn et al., 1977). Aerosol samples were collected by aircraft flying in haze bands located over the North Slope of Alaska. Physical and chemical measurements of these samples indicated that the strongest haze events were associated with natural dust particles. Subsequent meteorological analysis showed the dust to have originated from the great deserts of eastern Asia. These results supported the evidence of distant sources of dust and fresh-water diatoms from dry-lake beds found in snow water collected on the Arctic pack ice north of Barrow, Alaska during April, 1972 (Darby et al., 1974). A high-volume aerosol sampler was installed at Barrow, Alaska in an effort to determine the annual variability of the composition of the aerosol at that location. Subsequent results showed the aerosol to have a significant seasonal variation in concentration and composition and that there was a strong pollution aerosol component that peaked in the early spring. As a result, an informal organization of institutions with interest in arctic air chemistry was established to permit an integrated scientific effort on the study of Arctic Haze (Rahn, 1979). As more data on arctic air chemistry were brought together and compiled into one coherent picture, it became apparent that activities of man in mid-latitude regions had significant effects on the arctic aerosol chemical composition (Rahn and McCaffrey, 1980).

The overall results of the data pooled from the various institutions indicated that the winter arctic aerosol had all the characteristics of being well aged and derived from a polluted

continental source. This was confirmed by the use of chemical tracers such as noncrustal vanadium from combustion of heavy #6 residual oil; non-marine sulphate (from the oxidation of SO_2 emitted from the combustion of fossil fuels, primarily coal); the high concentrations of ^{210}Pb, a naturally occurring radionuclide derived from the decay of radon gas emitted from rock, which is indicative of the continental history of the air; and the grey color of filter samples, assumed to be a result of soot aerosol. The summer concentrations of these tracers ranged from 15-50 times less than the concentrations measured during winter. Based upon known sources, aerosol composition, meteorology of transport, and removal mechanisms, Rahn and McCaffrey (1980) argued that the source of the aerosol is consistent with a polluted European air mass transported over European Russia and then to the Arctic.

These findings spurred other investigators to study Arctic haze. Studies were undertaken on soot aerosol (Rosen et al., 1981; Heintzenburg, 1980) and organic material in the aerosol (Weschler, 1981; Daisey et al., 1981). These more detailed studies of the chemistry of the arctic aerosol supported the conclusion of an anthropogenic source which was deduced from the trace-elemental composition. Although aerosol mass loadings in the Arctic are low in comparison to mid-latitude regions, the relative composition of the aerosol, as shown by these studies, indicates the significance of remote sources of anthropogenic aerosol. Of the total carbon content of the arctic aerosol, up to 40% is in the form of graphitic carbon, a form of carbon specific to combustion sources. This proportion of graphitic carbon to the total carbon present is of the same order as that found in urban areas. Many of the other organic species present in the arctic aerosol

are strictly man-made. These purely man-made compounds provide further evidence that the winter arctic aerosol has a strong anthropogenic character. In addition, these results also show the summer arctic aerosol to be representative of a remote "background" region.

Anthropogenic, aerosol emissions of mid-latitude source regions do not have such a strong seasonal dependence. Several investigations of the meteorology of transport to the Arctic were conducted. The importance of upper tropospheric transport has been suggested by Reiter (1981) and Bigg (1980). The former is based upon an evaluation of planetary waves at the 500 mb level and their relation to vanadium-pollution episodes at Barrow, Alaska. The results indicate that the location and number of atmospheric planetary waves may be responsible for the long and short term variability of the arctic aerosol, but the study could not draw any conclusion as to what is the most significant source region for the aerosol. A study by Bigg (1980) suggested upper level transport may be important. He inferred a circulation cell beginning with vertical motion at low to mid latitudes, followed by slow northward movement resulting in subsidence at high latitudes. There is no evidence of such a circulation cell. The inferred circulation cell does not favor any particular region as a source of the aerosol.

Carlson (1981), using isentropic cross-sections of the atmosphere extending from mid-latitude source regions to the Arctic suggested that the source region must be north of the polar front. Based upon this requirement, he concludes that the most important source region is northeastern Europe and north central Asia.

Patterson and Husar (1981) constructed broad area trajectories at the 850 mb level from regions of strong pollutant emission. Their

results indicate that both European and North American source regions contribute to winter arctic haze, but are unlikely to contribute to the arctic aerosol during the summer. They comment that the major shortcoming of their model was the lack of precipitation scavenging of the aerosol and the assumed 850 mb isobaric flow for up to 20 days duration.

A unique approach to aerosol transport to the Arctic was considered by Shaw (1981). He developed an eddy-diffusion model to explain the observed concentrations of pollutant aerosol in the Arctic. The model ascribes the great seasonal difference to the effective removal of aerosol diffused to the north by the agglomeration of aerosol to the cloud droplets in the extensive summer arctic stratus clouds.

It becomes readily apparent from the above discussion that neither the arctic aerosol chemistry or atmospheric dynamics can explain the observed phenomenon of arctic haze individually. In order to fully understand the causes and significance of arctic haze, both approaches should be used in order to complement one another and provide a broad-based description of the problem.

2.2 Studies of the Arctic Cloud-Active Aerosol

Although there are a great number of studies of CCN and IN in the atmosphere, there are few that have been conducted in the Arctic. The area north of 60°N will be considered as the Arctic for the purposes of this discussion.

Probably the first attempt at determining which fraction of the aerosol was responsible for the formation of snow in the Arctic was a study conducted by Kumai and Francis (1962). During the summer of 1960, they collected snow crystals at Site 2, 320 km east of Thule, Greenland. The crystals were allowed to sublime on an electron microscope grid,

leaving behind the central nucleus. Their study showed that 85% of the nuclei identified were clay minerals. No combustion products were observed. They concluded that ice nuclei at this location during summer were primarily natural.

Aircraft measurements of CCN were made by Hopple et al. (1973) on the days of 15, 20, 21 and 22 February, 1972 at a location approximately 160 km north of Fairbanks, Alaska. In general, they found an increase in CCN concentrations with height. In particular they found the concentrations to sharply increase above the surface based temperature inversion. Their measurements also indicated that the CCN aloft were more volatile or smaller in size than those below the temperature inversion. This was done by measuring CCN from a stored sample volume and comparing the result to that determined by heating a portion of the same aerosol sample prior to activating the CCN. This would be consistent with aerosol having more of a sulphate-rich composition than being composed of marine or crustal based salts. At the time of their measurements, the winds above the temperature inversion were NW to NE, suggesting the CCN measured there were advected from the north.

During the months of February and March, 1968, measurements of IN concentrations active at $-15^{o}C$ were made near Fairbanks, Alaska at the surface. This was part of a large scale study of the significance of dust originating from the arid regions of eastern Asia as a source of IN over the North Pacific (Isono et al., 1971). They were able to show that IN "storms" detected at their Alaskan site could be correlated with dust storm events in the arid regions of North China and the transport of the dust over Japan, the North Pacific and on to western Washington state in the U.S.A. The IN were positively identified as soil

particles composed of clays and other minerals by electron microscope studies. Concentrations greater than 2 per liter were measured during these "storms". This occurrence is similar to that reported by Rahn et al. (1977) which showed the Asian dust can be transported to latitudes north of 70°N. However, further studies of the arctic aerosol (Rahn and McCaffrey, 1980) indicate that this flux of natural dusts is probably sporadic and not important to the overall average IN concentrations or aerosol composition in the Arctic.

At the same time (March, 1968) measurements of IN active at -17°C were being made by Higuchi and Wushiki (1970) at Barrow, Alaska using a filter technique. Concentrations of up to 5 per liter were measured when the filter was analyzed without warming above 0°C. Filters that were warmed above the freezing point usually showed a lower IN concentration, suggesting to the authors that a pre-activation mechanism operating in the atmosphere. Using isobaric trajectory analysis, they attributed the highest IN concentrations to air masses arriving from the Gobi desert regions of China where they suspected larger aerosol of terestrial origin could explain their findings. Their study is further evidence linking the great deserts of eastern Asia as a possible source region affecting the aerosol chemistry and nucleating properties of the atmosphere for thousands of kilometers downwind.

Laktionov (1969) made measurements by aircraft of CCN concentrations and spectra near the cities of Kiev (51°N, 31°E) and Archangelsk (64°N, 41°E) during July, 1968. He reports CCN concentrations decrease by a factor of 30-100 from an elevation of 200 m to 4000 m over a range of supersaturations from 0.1% to 1.6%. These measurements appear to show a surface source of CCN over these

continental regions during the summer. The results of Laktionov are in agreement with the observations of Twomey (1963) and Squires (1966) for populated continental regions. The concentrations of CCN at Kiev ranged from 50% to 500% higher than those at Archangelsk as measured at cloud base. These data suggest that as one approaches the Arctic Ocean coast in summer, the concentrations of CCN decrease from those determined at more continental locations. The concentrations of CCN over the entire range of supersaturations >0.3% reported at Archangelsk during the summer are less than those reported by Hopple et al. (1973). These data support other evidence of a continental or anthropogenic source of CCN and the relatively higher concentrations of aerosol in Arctic regions during the winter vs. that of the summer.

Radke et al. (1976) reported on the concentrations of both CCN and IN at Barrow, Alaska during the period 8-28 March, 1970. Measurements of CCN concentrations were, on the average, typical of those measured at other similar locations. They also reported CCN concentrations ranging from the lowest ever recorded to values that were representative of urban continental air masses. They found maxima in CCN concentrations to correspond to winds from the northeast at the surface, the clean air sector, and minima when winds were from the direction of the closest local aerosol contamination source, the village of Barrow. They were unable to explain the high CCN concentrations by stratospheric intrusions of sulphate aerosol. Their final explanation was based upon either transport of aerosol from sources near the Deadhorse - Prudhoe Bay oil field development or from long range transport from northeastern North America.

The measurements of IN at -20°C by Radke et al. (1976) ranged over three orders of magnitude. The average and geometric mean were 0.5 per liter and 0.02 per liter respectively, indicating the persistance of low concentrations. A wind rose for the IN concentrations showed the highest concentrations occurred when winds were from the NE to NW. Minimum IN concentrations occurred with winds from the SE. They concluded there were different sources for the IN and CCN.

Jayaweera and Ohtake (1973) determined the relationship between IN measured at cloud top temperature and the number of ice crystals present in arctic stratus clouds. Although the IN were measured at the surface, they found excellent agreement between the number of IN and the observed number of ice crystals present in stratus clouds overhead for the temperature range -4°C to -20°C. These measurements were made in September, 1971 and April, 1972 and are within the range of values determined at two sites near Fairbanks, Alaska over the period January, February and March, 1969 (Bigg and Stevenson, 1970). Their reported concentrations of IN at -20°C were 0.4 - 0.9 per liter, comparable to those values reported by Radke et al. (1976).

Most recently, measurements of CCN and IN were made from the instrumented National Center for Atmospheric Research (NCAR) Electra aircraft during the month of June, 1980 (Saxena, 1981; Ohtake, 1981; Jayaweera and Flanagan, 1981). Measurements of CCN showed increases in concentration from the surface to 2000 m, concentrations up to 4000 cm^{-3} in evaporated cloud air for the supersaturation range $0.1\% < S < 1.0\%$ and that CCN were composed primarily of sulphur containing compounds. Biogenic IN were also measured. Two bacteria and one fungal spore were found to act as IN. Concentrations of these biogenic IN were shown to

be 1 m^{-3} at a temperature of $-6^{o}C$. It was suggested that much of the IN active at these warm temperatures may be due to biogenic particles in the atmosphere. These concentrations of IN and the vertical distribution of CCN are in agreement with those of Bigg and Stevenson (1970) and Bigg (1980).

2.3 Cloud Microphysical Studies in the Arctic

There are few reported studies of the microphysical properties of arctic clouds. Although it is certainly valid to generalize the known properties of cloud types from mid-latitude regions and extrapolate them to the Arctic as a first estimate of what should be expected, based upon the recent finding that the arctic aerosol is significantly pollution-derived, this should be done with extreme caution. The data from the few investigations of arctic clouds are summarized. As in the case of the arctic cloud-active aerosol, the region north of $60^{o}N$ will be considered as the Arctic.

An extensive study of arctic fog, stratus and stratocumulus was reported by Gergach et al. (1960) over the Kara Sea east of Novaya Zemyla during the fall of 1956. Median drop radii and range of drop radii were reported. Unfortunately, no drop size distributions were reported. In general, the values were typical of those reported by other investigators in other regions for the same cloud type (Pruppacher and Klett, 1980). The vertical thickness of stratus was usually 150-250 m with the range being 50-700 m. The range of thicknesses of stratocumulus clouds was 150-600 m. Mean droplet radii reported for stratus clouds was 7-10 μm. Stratocumulus clouds had a broader drop size distribution. Maximum sizes sometimes exceeded 100-200 μm in radius.

A more detailed study of arctic stratus clouds was conducted during early September, 1959 by Koptev and Voskresenkiy (1962). Aircraft flights were conducted along the 125°E longitude line as far north as 83°N. They reported cloud liquid water contents (LWC) ranging from 0.06 to 0.15 gm m^{-3}. The droplet modal size ranged from 4.9 to 7.1 μm, radius. The larger modal size and broadness of the size distribution occurred in the clouds with higher LWC. Droplet concentrations ranged from about 18 to 91 cm^{-3}. Typically 40% to 60% of the droplets occurred in the range 2.5 to 5.0 μm radius. They also showed that the albedo of the cloud was positively correlated, in part, to the numbers of small cloud droplets present. No assessment was made as to the causes of the observed differences in the cloud microstructures.

Other Soviet studies reported by Laktionov (1969) compared the CCN spectra below cloud base with observed cloud droplet size distributions in small cumuli near the city of Archangelsk at 64°N during July, 1968. The measurement technique did not allow the droplets smaller than 5 μm diameter to be counted. However, the distributions are typical for those of small cumuli over continental areas where the number maxima occurs at a size of approximately 5-7 μm diameter with the total number concentration being around several hundred per cubic centimeter (Pruppacher and Klett, 1980). Their results indicate that there may be a general decrease in droplet numbers as one moves further north from more continental regions of European Russia toward the Arctic Ocean near Archangelsk. This was also confirmed by their measurements of the CCN spectra as discussed in section 2.2 previously.

The first investigation of winter arctic stratus cloud microphysics undertaken in the American Arctic was reported by Witte (1968). Several

measurements of cloud droplet size distribution, ice crystal size distribution, and LWC were conducted from aircraft at the Naval Arctic Research Laboratories, Barrow, Alaska. The measurements were made at altitudes ranging from 150 m to 3.54 km and temperatures ranging from $-19^{o}C$ to $-34^{o}C$. The measurements were made during the period 2-16 December, 1967. The measurement technique precluded any droplet or crystal measurements for sizes less than 2 μm radius. The droplet size distributions were all narrow with the majority of the droplets less than 10 μm radius. Witte suggested that these narrow distributions indicate that droplet coalescence processes are not important in Arctic clouds. He also concluded that droplet growth by diffusion dominates and that updrafts in the clouds were very uniform. Droplet number concentrations ranged from 10-50 cm^{-3} with liquid water contents from 0.0005-0.069 gm cm^{-3}. There is some indication from the data presented that maxima in droplet concentrations occur at intermediate levels i. e. 1.5-2.5 km altitude. This is from a small data set and may not be indicative of a true average. These results indicate arctic stratus cloud have one of the lowest LWC and smallest droplet concentrations of any cloud type.

Ice crystal concentrations were relatively high. Concentrations ranged from 200-1100 per liter over the observed cloud temperature range of $-19^{o}C$ to $-34^{o}C$. No relationship was observed between cloud temperature and ice crystal concentration. One might expect some form of ice-multiplication process at work, however the criteria for such a process as described by many authors (Pruppacher and Klett, 1980) requires warm temperatures and high liquid water contents in the clouds. Using the relationship of Fletcher (1962) for the global average IN

concentrations versus temperature, a temperature of at least -30°C is required to explain the observed crystal concentrations. Two possible explanations are: 1) based upon the microphysics of the clouds and the dominance of the diffusional growth process, the only removal process is sedimentation or evaporation. Witte (1968) estimates the lifetime of an arctic stratus cloud droplet could be approximately 11 hours. This is generally an order of magnitude greater than that which may occur in a cloud where LWC is high, ice multiplication occurs, and there is a greater amount of turbulence, all of which enhances the coalescence process and reduces droplet lifetimes. Since ice nucleation is a kinetic process, with a given nucleation rate for every combination of in situ conditions, the more time allowed for a nucleational event to occur (the freezing of a droplet heterogeneously, for instance) the greater the total number of possible nucleational events observed (the formation of an ice crystal). The observed number of ice crystals may be a unique combination of time, temperature and available IN that occur during the winter in arctic stratus clouds. 2) From the observations of Witte (1968), although small in number, it is apparent that a simple relationship of crystal concentration with temperature or altitude is not possible. The observed crystal concentrations may be due to the different nucleational properties of the ambient aerosol present and its vertical distribution at the time of the observation. This is only speculation since IN concentrations were not determined. Simultaneous measurements by others (Auer et al., 1969 and Hobbs, 1969) of IN and ice crystal concentrations showed that for temperatures lower than -26°C the ice cyrstal concentrations correspond to the IN concentrations, especially for stable stratiform clouds. The high ice-crystal

concentration observed by Witte (1968) are probably due to some combination of the two explanations given above and other effects not considered here.

A study similar to the one discussed above was conducted by Jayaweera and Ohtake (1973) using aircraft to sample cloud particles near Barrow, Alaska during the months of September, 1971 and April, 1972. They concluded from the microstructure of the arctic stratus clouds that they were colloidally stable. However, there are basic differences between their reported cloud microstructure and that reported by Witte (1968). The primary differences were higher LWC, drop size distributions skewed toward larger sizes, higher droplet concentrations, and lower ice crystal concentrations. Their measurements did not allow collection of cloud particles less than 4 μm radius. The overall average droplet size distribution had a mean radius of 7 μm and a range of 4–17 μm, again a relatively narrow distribution. Average droplet concentrations were 90 cm^{-3}, while the highest concentration was 160 cm^{-3}. LWC was typically in the range 0.1–0.2 gm m^{-3}. The most likely reasons for the differences between the observations of Witte (1968) and those of Jayaweera and Ohtake (1973) were differences in environmental conditions. Jayaweera and Ohtake (1973) conducted their investigation primarily during the arctic summer. This is demonstrated by the range of cloud temperatures they observed, $0^{o}C$ to $-18^{o}C$. These are warmer than any of the temperatures reported by Witte (1968). The temperature difference is also apparent in the ice crystal concentrations in these warmer clouds. They report values ranging from 0.002 l^{-1} (their lower limit of detection) to 0.21 l^{-1}. These values are 10^{3} to 10^{6} lower than those of Witte (1968). Their

observations are supported by simultaneous ground based measurements of IN concentrations at $-15^{o}C$ and $-20^{o}C$ and by observations of others done at two other sites in Alaska (Bigg and Stevenson, 1970), however the latter sites were in the Fairbanks, Alaska area. Jayaweera and Ohtake concluded: 1) there are negligible concentrations of ice crystals in comparison to droplets in the summer arctic stratus. 2) Ice crystal riming is of little to no significance and thus may be the single most important factor for the lack of ice crystal multiplication. 3) The radiative properties of these clouds is controlled by the droplet size spectrum and concentration, and not by the ice crystals.

There have been reports of rimed ice crystals in the Arctic. In a collection of reports edited by Magono (1978) numerous observations of rimed snow and grauple are presented. These observations were made during January and February, 1977 at Inuvik, N.W.T., Canada ($68^{o}N$, $134^{o}W$) at the surface. Although surface temperatures were typically $-20^{o}C$ to $-40^{o}C$, they noted temperatures between the 850 mb and 700 mb pressure levels were frequently near the freezing point. These conditions were usually due to warm, moist, North Pacific air from the southwest over-riding the surface based temperature inversion; an intrusion of non-Arctic air masses from the south. Several investigators were able to derive cloud droplet size frequency distributions by studying the droplets intercepted by falling ice crystals. These data show drop distributions with a size ranging from 5 μm to 40 μm radius and mean radii of about 19 μm. These data must be interpreted with caution since the collection efficiency of ice crystals is very poor for droplets with radii $<$ 10 μm (Pruppacher and Klett, 1980), and the clouds were probably high in LWC and had broad droplet

size distributions more representative of the warmer maritime conditions of the air mass source, the North Pacific.

Finally, studies of arctic fog will be briefly discussed. Kumai (1965 and 1973) investigated arctic fog microstructure and the origin of fog nuclei. He found values of LWC to range from 0.002 to 0.09 gm m^{-3} and drop size to range from 2–140 μm radius. Droplet concentrations ranged from 0.02–0.63 cm^{-3}. All of his observations were made during the summer of 1971 at Barrow, Alaska. All measurements were made during clean "background" conditions as confirmed by the Aitken nuclei concentrations. Studies of the nuclei of the fog droplets by electron microscope and diffraction techniques showed that 91% of the nuclei were of sea salt origin. The remainder were too small to identify. In general, the observations of LWC and size are typical for fogs. The lower droplet concentrations reported in comparison to those of others (Pruppacher and Klett, 1980) reflect the inability of his collection technique to measure droplets less than 2 μm radius, where the majority of fog droplet numbers occur. The region surrounding Barrow, Alaska is maritime in character during the summer, but due to the fact that much of the Arctic Ocean is still covered by ice, limiting the fetch of the wind over open water, and the low wind speeds reported at the time of the observations, it is expected that the production of sea salt particles would be inhibited.

In summary, measurements of Arctic cloud microphysics are limited in number and it is difficult to deduce much about the general microstructure of the clouds of this region. Arctic clouds are important to the regional budget and the global climate. In the future more coherent

and multidisciplinary studies are needed, during all seasons of the year, to properly characterize Arctic clouds.

III. STATEMENT OF THE PROBLEM AND OBJECTIVES

3.1 The Problem

3.11 Arctic aerosol and its relation to Arctic clouds

Despite the recent interest in the aerosol in Arctic regions, the primary thrust of most investigations has been toward the chemical composition of the aerosol. These chemical measurements were the key to understanding how important imported aerosol was to the overall aerosol composition. No studies have been concerned with the dependence of the nucleational characteristics of the aerosol on the natural and anthropogenic components. It is generally believed that one of the most important climatic effects of aerosols in the atmosphere is on the radiation budget. A few studies have been concerned with the effects of the aerosol on turbidity, light scattering and absorption (Shaw, 1975; Bodhaine et a., 1981; Heintzenberg, 1982). On global and regional scales, clouds are the single most important factor in determining the radiative properties of the atmosphere. In the Arctic the persistent stratus clouds are one of the major atmospheric components of the radiation balance (Fletcher, 1965). The interaction of clouds with the radiation field depends upon the cloud microstructure (Welch and Cox, 1980; Twomey, 1977). The transmissivity and reflectivity of clouds depends upon the cloud droplet size distribution and concentration. High droplet concentrations result in higher albedos and lower transmissivities of clouds. Increase in small sized ice crystals (<100 major dimension) cause increases in cloud reflectance and absorptance.

Changes in the microstructure of clouds can be related to the concentrations of CCN and IN in the air mass from which the clouds evolve (Twomey, 1959; Fitzgerald and Spyers-Duran, 1973; Hobbs et al., 1980). Measurements of CCN and IN in the Arctic, and the determination of their sources, would be valuable in assessing the effects of anthropogenic aerosol on the radiative properties of arctic clouds. This study addresses the nucleational aspect of Arctic Haze for the first time. This is important in the overall understanding of the effects of the haze aerosol because the nucleational properties of the aerosol depend upon the physical and chemical properties of the dry aerosol.

3.12 CCN and IN - sources for the Arctic

The chemistry of the Arctic aerosol has revealed a great deal about which aerosol sources play a major role for this region. The question being addressed with regard to the cloud-active aerosol in the Arctic is how significant the man-made sources are. The aerosol chemistry shows a strong seasonal dependence of natural and anthropogenic components present in the aerosol. It is logical to expect that any man-made sources of CCN and IN in the regions suspected of being sources for the bulk Arctic aerosol would be significant sources of CCN and IN for the Arctic as well.

Many investigations of anthropogenic sources of CCN and IN have been conducted in the past (Schaefer, 1969; Weickmann, 1972; Hobbs et al., 1970; Van Valin et al., 1975, 1976; Van Valin and Pueschel, 1976; Parungo et al., 1978; Pueschel et al., 1979; etc.). It is not possible to simply extrapolate from concentrations of cloud-active aerosol at mid-latitude source regions to that expected in the Arctic, based upon

measured concentrations of various pollutants. There are at least three reasons for this: 1) The number of CCN present in a pollution plume depends in part upon the age of the plume due to gas-to-particle conversion of SO_2 to sulphate aerosol (Hobbs et al., 1980; Meagher, et al., 1978; Mamane and Pueschel, 1980). Interaction of IN, both natural and man-made, with other chemical species in a pollutant plume may decrease the number of IN (Georgii and Kleinjung, 1967; Braham and Spyers-Duran, 1974). Thus the net effect of a pollutant plume may be either positive or negative with regard to IN concentrations. 2) Any pollutant aerosol transported to the Arctic must travel over several thousand kilometers. All along the transport path will be sources of natural IN and CCN. 3) Because of the nature of these nuclei, they participate in the precipitation-formation process. Thus they would be highly susceptible to removal from the atmosphere if precipitation occurs along the transport path. It is therefore necessary to measure these nuclei in the Arctic in order to assess the net effects of these various input and removal mechanisms.

Cloud-active aerosol from natural sources can also be transported to the Arctic. Some fraction of the CCN and IN is probably generated within the Arctic. Considering that long-range-transport of aerosol to the Arctic is well-documented, certainly any of the known natural sources of CCN and IN may play a role in the Arctic. CCN are known to exist in higher concentrations over continents than over oceans (Twomey and Wojciechowski, 1969). In general, these nuclei are most likely ammonium sulphate (Dinger et al., 1970) from ammonia and sulphur gases from by biogenic sources. Although sea-salt particles would be expected to be a significant component of the marine sources of CCN, direct

measurements of the sea-salt salt particle concentration indicate that only 10% could be sea-salt. Recent investigations suggest that biogenic material from the oceans may be a significant source of CCN in marine areas (Saxena and Baier, 1981). Another natural source of CCN is combustion products from forest fires (Hobbs and Radke, 1969; Eagan et al., 1974). Natural sources of IN have been shown to be some mixture of soil or clay particles (Mason and Mayband, 1958; Isono et al., 1959), possibly meteoric material (Mason, 1960; Bigg and Giutronich, 1967), volcanic emissions (Isono et al., 1959), the oceans (Brier and Klien, 1959) or various biogenic particles (Jayaweera and Flanagan, 1981; Schnell et al., 1981).

Many of these natural sources of nuclei are sporadic in time and space and are not significant in the mean. Other sources of large geographic extent must be taken into account as viable inputs to the total nuclei concentration present in the Arctic.

3.13 Response of Arctic clouds to the aerosol

Arctic cloud climatology is dominated by stratiform clouds. Because of the near lack of aerosol sources within the region, as a result of little human activity and the presence of a snow and ice cover much of the year, the Arctic can be viewed as a relatively simple climatic system. Stratiform clouds, because of the small amount of turbulence associated with them, have drop size distributions that can be predicted from the CCN spectra. Hindman et al. (1977) showed that more complex small cumuliform cloud droplet size distributions could be predicted from CCN spectra. The Arctic stratus form in a stable atmosphere. Differential updrafts in other clouds are generally believed to be responsible for broadening droplet size distributions.

Thus the stratus cloud droplet size distributions of an Arctic stratus cloud should depend only upon the CCN spectra and the diffusional growth process. This is apparently a valid argument, based upon the measurements of Vitte (1968) discussed previously. In addition, Auer et al. (1969) and Hobbs (1969) have shown that for temperatures colder than $-26^{o}C$, ice-crystal concentrations can be predicted from the IN concentration, especially in stratiform clouds. It is apparent that measurements of CCN and IN in the Arctic should be useful as a predictor of cloud microstructure. Therefore, any changes in IN and CCN concentrations as a result of man's activity should be reflected in the cloud microstructure. This places a great deal of significance upon the transport of mid-latitude pollutants to the Arctic and the understanding of their nucleational properties.

3.2 Objectives

The primary goal of this study is to assess the role of imported anthropogenic aerosol in the Arctic on the concentration of CCN and IN. Multiple sites around the perimeter of the Arctic were chosen to insure, as much as possible, that the arctic studies are representative of the regional nuclei concentrations. Differences in seasonal averages, and day-to-day concentrations are interpreted based upon the large-scale controlling meteorological factors of long-range transport, and the smaller-scale synoptic weather patterns of the region respectively. This is done to ensure that the measurements are representative in time. The aerosol trace-constituent composition, determined simultaneously with the nucleus measurements, is used to assess the relative influence of man-made and natural aerosols present in a given airmass under study.

The measurements of the physical and chemical properties of the arctic aerosol are then interpreted using simple meteorological analysis. Combining this with knowledge of the general circulation of the atmosphere, the second goal, to assess what major industrial regions of the Northern Hemisphere are contributing to the presence of pollutants in the Arctic, is addressed. As a result, the third goal, determining atmospheric pathways of pollutants to the Arctic, is discussed in light of the chemical and meteorological studies.

Finally, the fourth goal is to address the possible effects of pollutants transported to the Arctic on the microstructure of Arctic clouds. The discussion will center on what changes may be expected in the droplet and ice crystal number concentrations in Arctic clouds. The conclusions will be based upon the results of the measurements of changes in CCN and IN concentrations, respectively and how they, in turn, are related to the change in pollutant aerosol concentrations. The results can then be used to evaluate whether effects on Arctic cloud microstructure are significant and in need of further study.

IV. EXPERIMENTAL DESIGN

Prior knowledge of arctic aerosol chemistry, mean circulation patterns of the atmosphere and major suspected pollution aerosol source regions determined the framework for the design of the field program. Combining this with available facilities, the field-work design evolved. All sample collections and measurements used standard techniques. The end result was a set of measurements that were unique in that they brought together chemical and physical measurements of the arctic aerosol to allow a broader base for interpretation.

4.1 Overall Perspective

The experimental design was based upon chemical and physical measurements of the arctic aerosol at various locations in the Arctic. The locations were chosen to provide a maximum amount of information on possible long-range transport of aerosol to the Arctic. All sites were visited both during winter and summer, because of the seasonal dependence of the chemistry of the arctic aerosol. During each site visit, collections and measurements of the aerosol were made over 7-11-day periods. This was felt to be a minimum length of time to ascertain the daily variability of the aerosol due to changes in the synoptic weather patterns.

Aerosol-particle studies were the same at all sites visited. Analysis methods used were the same for all samples from all sites. This was intended to keep the number of variables pertaining to the aerosol measurements from site to site to a minimum. Details of the

sites and the methods used for aerosol collection and analysis will be discussed in the following sections.

4.2 Arctic Field Sites and Their Justification

Figure 4.1 shows likely pathways of atmospheric transport of aerosol to the Arctic. This diagram was compiled from basic knowledge of the general circulation and known regions of sources of pollution of aerosol (Rahn and McCaffrey, 1980). It is important to be aware that this does not reflect any removal processes that may characteristically occur along these pathways. Modeling studies by others also support this picture.

The solid line in Figure 4.2 depicts the mean relative topography along 50°N of the normal pressure surface maps of January given as departures from the mean value of the layer of the atmosphere between 100 and 1000 mb (Reiter, 1969). This figure shows the importance of a two-wave regime, primarily controlled by the cold Asian and North American troughs. The longitudinal location of these troughs is given by the valleys in the graph. The longitudinal regions of the plot where the departure of relative topography is increasing, are regions of persistent transport to the north. These regions are 30°-90° W, 140° E-150° W and a region near 75° E. The two oceanic regions correspond to the two major areas of cyclogenesis or eddy development in the Northern Hemisphere: the Northern Atlantic and Pacific Oceans. The region near 75° E, suggested in Figure 4.2 by a flattening of the curve, appears as the arrows in Figure 4.1 showing transport over central Siberia, and some portion of the arrow with origins in Eastern Europe. This is a region of weak northerly transport and is not an area of cyclogenesis.

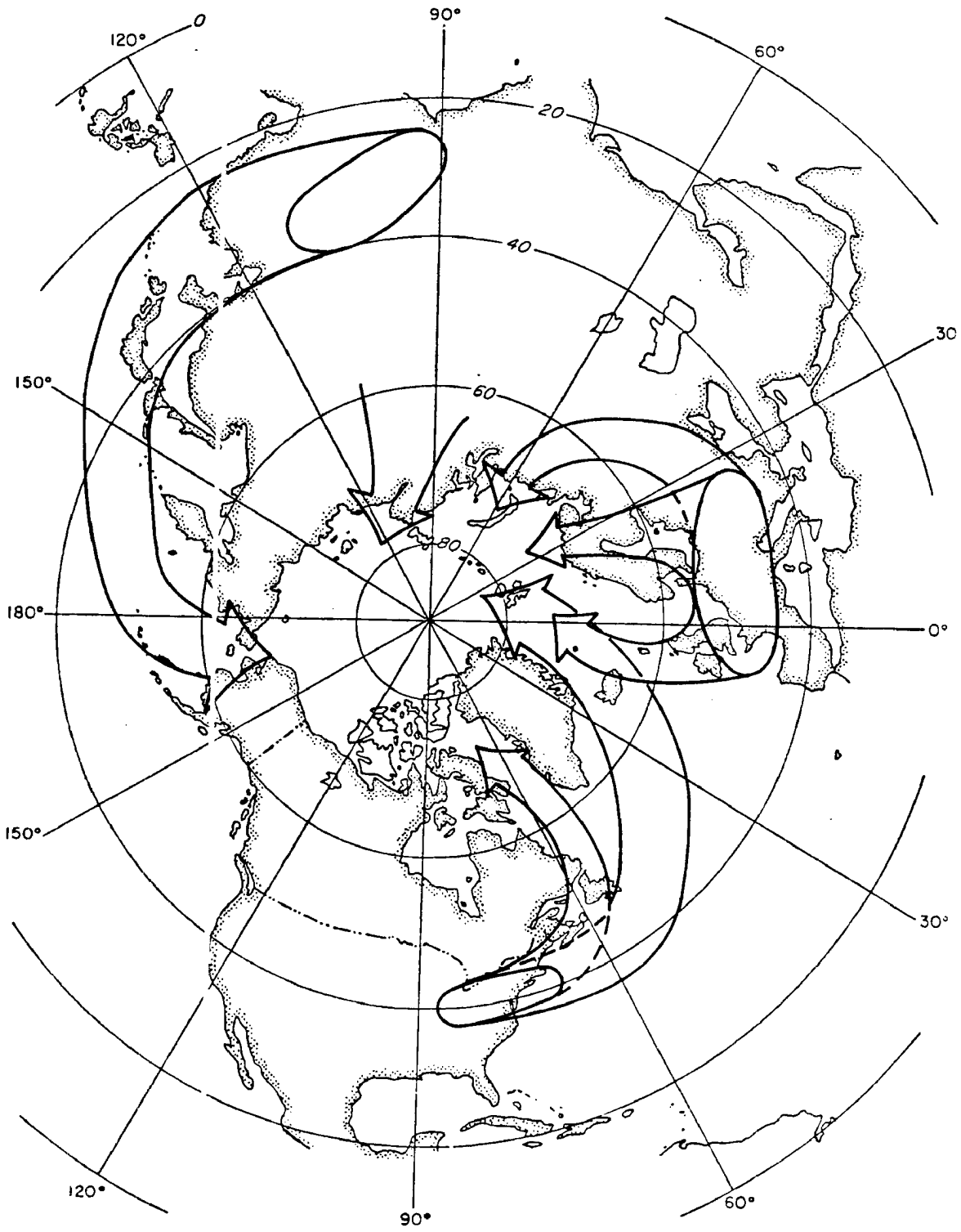


Fig. 4.1 A schematic depiction of proposed pathways of long-range atmospheric transport of pollutants to the Arctic based upon climatic data on mean meridional transport, preferred storm tracks and known major source regions of pollution aerosol.

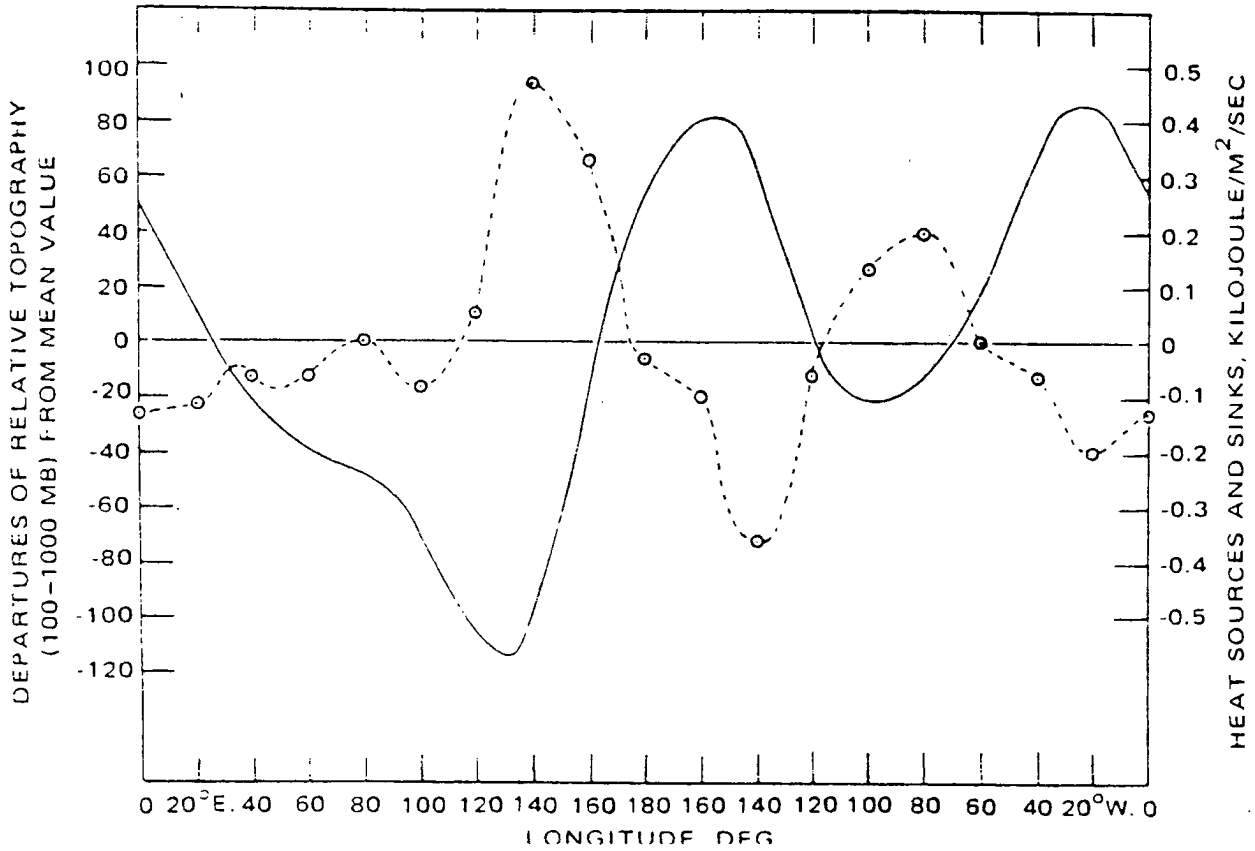


Fig. 4.2 Mean relative topography (solid line) along 50° N for the normal maps of January, as departures from the mean value of the layer of the atmosphere between 100 and 1000 mb (after Reiter, 1969).

There are basically two major ways to transport air to the Arctic, mean transport due to the persistent two-wave pattern of the atmospheric planetary-wave regime, and transport via eddies on the mean. Figure 4.3 shows a tentative model of the importance of mean meridional versus eddy flux of energy in the form of sensible heat plus potential energy in the Northern Hemisphere for summer and winter (Reiter, 1969, pp. 98-99). It is apparent that eddy transport dominates the mean meridional transport, and is extremely important in the winter. The eddy-energy transport is associated with cyclonic activity.

Petterssen (1958) summarized the eddy transport paths in the Northern Hemisphere as follows: "On the Pacific side, we note a broad band of high cyclone activity extending from Southeast Asia to the Gulf of Alaska. Most of these cyclones form on the Pacific polar fronts. . . During the cold season, these storms acquire great intensity. Most of them travel northeastward and accumulate in the Gulf of Alaska, and this accounts for the high frequency (of storms) in this region. On the Atlantic side, the conditions are similar to those on the Pacific Ocean. Storms develop frequently on the Atlantic polar front. The most favored region is the coast of Virginia and the general area to the east of the southern Appalachians. These move more or less along the Gulf Stream, develop to great intensity, and tend to stagnate in the vicinity of Iceland or over the waters between Greenland and Labrador. Many cyclones form or redevelop on the Atlantic arctic front and move eastward to the Barents Sea, as far east as Novaya Zemyla. Some of them continue along the entire coast of Siberia."

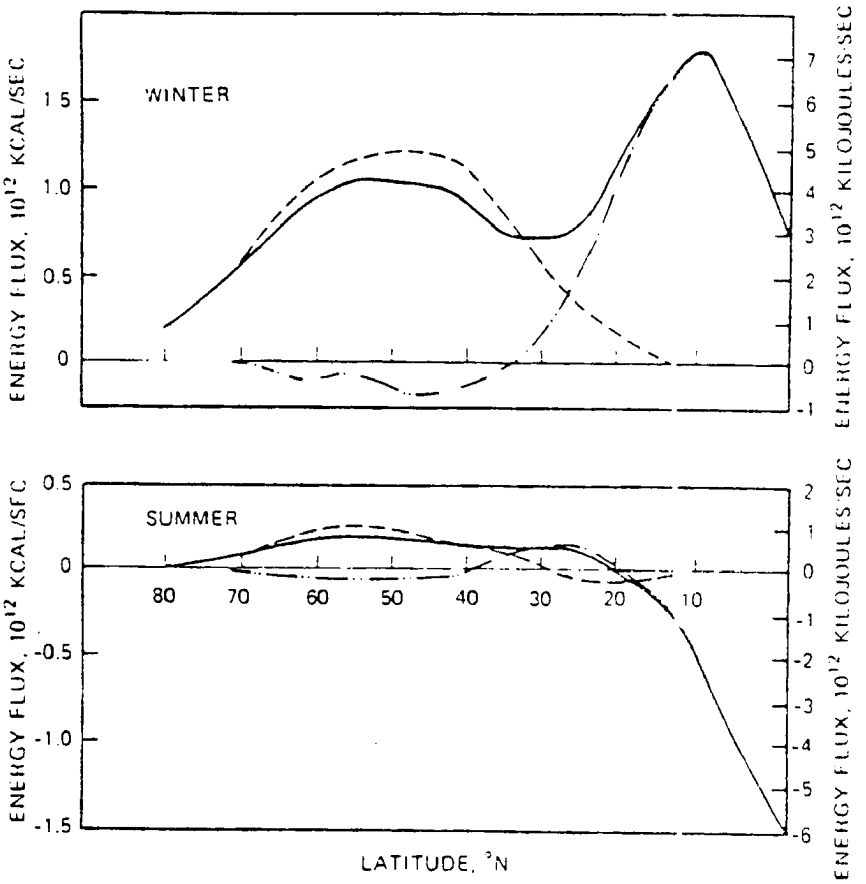


Fig. 4.3 Mean poleward energy flux for the northern hemisphere atmosphere (after Reiter, 1969). — = total flux, —··— = flux associated with mean meridional circulation, ------ = flux associated with zonal eddies.

The best locations from which to investigate the effects of the long range winter transport of aerosol to the Arctic would be (1) in or downwind of major aerosol source regions, (2) along the eddy-transport path, (3) at the final or endpoint of the transport path to the north. From the locations of the industrialized nations in the Northern Hemisphere and the transport pathways shown here in figure 4.1, the most effective gateway to the Arctic for pollutants would be the North Atlantic sector. Summer field work performed at the same sites would permit the evaluation of the differences between poleward aerosol transport of summer and winter, and how it effects the concentration of the cloud-active aerosol at each location.

As a result, five Arctic and one mid-latitude sites were chosen: Barrow, Alaska; Poker Flat, Alaska; Panguirtung, Canada; Irafoss, Iceland; Ny Alsund, Spitsbergen; and Narragansett, Rhode Island. Location of these sites are shown in Figure 4.4. Table 4.1 summarizes the dates each site was visited, and the number of sample sets collected. The site at Ny Alesund was not revisited during the summer season. Instead, a cruise on the Swedish icebreaker expedition YMER-80 was substituted. The cruise took place in the region of Spitsbergen and Greenland. A brief discussion of each of the sites and their justification follows.

4.2.1 Barrow, Alaska, USA

Near the village of Barrow, Alaska (71° N, 156° W), the combined facilities of the Naval Arctic Research Laboratory (NARL) and the Global Monitoring for Climatic Change (GMCC) station operated by the National Oceanic and Atmospheric Administration (NOAA) provided an excellent

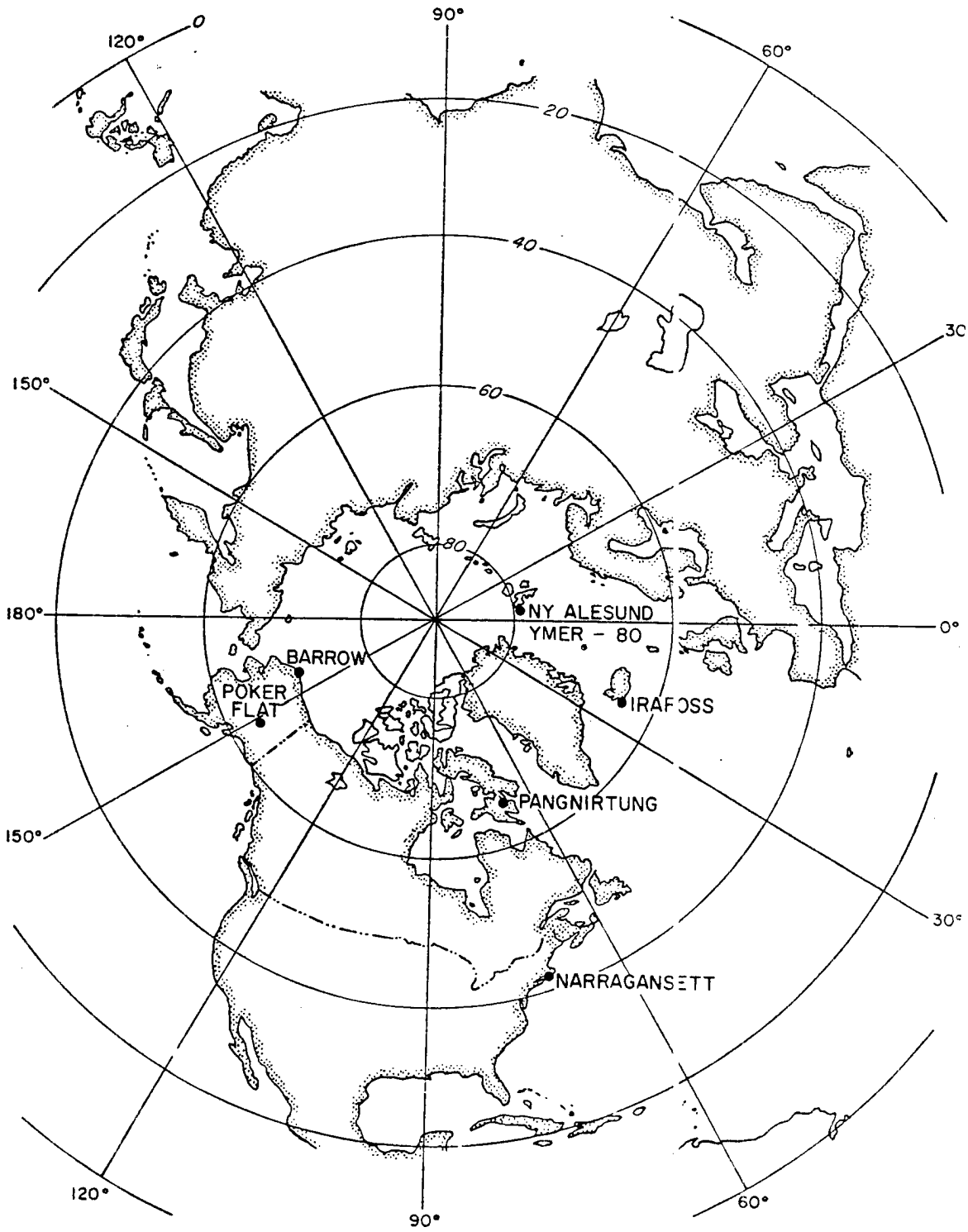


Fig. 4.4 The locations of the five arctic aerosol experiment sites and one mid-latitude source region site.

TABLE 4.1
Summary of field studies of cloud-active aerosol in the Arctic and environs.

Site	Dates	Number of samples sets
Barrow, AK	27 July - 7 August 1979	10
	23 March - 3 April 1980	10
Poker Flat, AK	17 - 25 July 1979	9
	4 - 13 April 1980	10
Pangnirtung, NWT, Canada	21 June - 30 June 1979	9
	26 January - 3 February 1980	7
Irafoss, Iceland	14 - 25 August 1979	9
	16 - 23 February 1980	7
YMER-80	11 August - 1 September 1980	11
Ny Alesund, Spitsbergen	28 February - 11 March 1980	11
Narragansett, RI	12 - 19 June 1979	7
	6 - 15 January 1980	10

location for study of the arctic cloud-active aerosol. All measurements and collections were made at the GMCC facility located upwind of the major human activity in the village at Barrow and NAIL.

The prevailing wind direction is northeasterly at this site. This provides the site with air arriving from the Arctic Ocean basin to the north. Detailed studies of the aerosol at this site by others (Bigg, 1980; Rahn and McCaffrey, 1980; Rahn, personal communication) have shown the aerosol to be representative of the background regional arctic aerosol. In addition, previous research has shown that airmasses from the south can penetrate as far north as Barrow (Rahn et al., 1977). Thus from the standpoint of long range transport, Barrow lies along the North Pacific pathway as well as the "end of the line" of potential aerosol transport from the North Atlantic pathways.

4.2.2 Poker Flat, Alaska, USA

A second Alaskan site was chosen at the Poker Flat Research Range (65^{o} N, 147^{o} W) operated by the University of Alaska. This site represents interior Alaska. It lies between Barrow and potential transport along the North Pacific pathway. Thus it is a good location from which to study the effects of aerosol transport to the north. This is especially true during the winter when the interior is snow-covered, eliminating much of the locally derived natural aerosol, dusts and biogenic material. Airmasses can reach this location from both the north or south. This also allows the arctic airmasses to be contrasted with those arriving from the south. Past experience has shown the Brooks Range, lying between Barrow and Poker Flat, effectively isolates the two sites both meteorologically and chemically, except for occasional intrusions. Thus, comparisons of the two sites allows the

North Pacific pathway to be assessed in terms of aerosol transport to the north from remote regions (Asia and the ocean) and the Alaskan interior. This site is located approximately 25 km north of the city of Fairbanks at an elevation of 490 m, about 340 m higher than the city.

4.2.3 Pangnirtung, NWT, Canada

Pangnirtung is a small village (pop. 800) located on the SE end of Baffin Island. This site was chosen to lie along a potential pathway mid-way between Barrow and the northeastern United States (see Figure 1). This is the shortest possible route of mid-latitude pollutants to the North Slope of Alaska (Barrow). This route was first suggested by Radke, et al. (1976) as a possible explanation of their observed elevated concentrations of CCN at Barrow. Because of the lack of any ongoing aerosol measurement program in this area, sampling was done from a tent located upwind of the village, on the shore of a point of land extending into a fjord. This proved satisfactory since air flow past the sampling site was controlled by the fjord. The prevailing wind direction was either up- or down-fjord, effectively excluding the village.

4.2.4 Irafoss, Iceland

This site was located at a hydroelectric station approximately 35 km east of Reykjyvik, Iceland. The area has few inhabitants. Most of the energy used in Iceland is derived from geothermal or hydroelectric sources. This makes the site an excellent one for the study of long range pollution transport. Iceland is located along the pathway originating in the northeast United States. In addition, it can also be influenced by transport from Europe via a retrograde easterly flow which can be associated with either high or low pressure centers between

Iceland and Europe. This allows a possible comparison of the relative strengths of pollution aerosol advection from Europe and the United States to be compared. These two regions have the largest pollutant emissions in the Northern Hemisphere. This site in the mid-Atlantic offers the possibility of studying the net effect of long range pollutant transport associated with these two regions on the cloud-active aerosol in Iceland.

4.2.5 Ny Alesund-YMER

The sampling location at Ny Alesund, Spitsbergen (79°N, 12°E) was only visited during the winter season. The summer portion of the field experiment took place at sea near Spitsbergen and Greenland. The summer experiment was conducted onboard the H. M. S. Ymer, a Swedish icebreaker outfitted for scientific studies in the Arctic during the summer of 1980. The area around Spitsbergen has several reasons for being well suited for aerosol studies. 1) It has had a continuous aerosol sampling program operated by the Norwegian Institute for Air Research (NILU) located at Ny Alesund for many years. This provides a background of information for the site and an existing air sampling facility. 2) It lies along the proposed transport paths from North America and Europe. Thus it provides a unique location from which to determine the differences in the aerosol being advected from these two regions to the north. 3) At 79° N it is well within the Arctic and will also be affected by polar air masses. In all, these features serve to provide a wealth of information from one location.

4.3 Narragansett, Rhode Island, USA - Source Region

This site was chosen for two reasons: 1) The existing aerosol sampling station and analysis facilities at the University of Rhode

Island, Graduate School of Oceanography and 2) the location of the site within a possible major source region of anthropogenic aerosol for the Arctic, the northeastern United States. For the latter reason, it is important to perform the same aerosol experiments in or near this source region as those done in the Arctic. This site is used to determine the nature of the cloud-active aerosol fresh from a strong source region suspected to be representative of the anthropogenic aerosol affecting the Arctic.

4.4 Aerosol Experiments Conducted

4.4.1 General Collection Strategy

The overall objective at each sampling site was to collect aerosol samples, both high-volume for chemical analysis and low-volume for physical studies, to be used to characterize the aerosol and air mass as to the relative proportions of natural and anthropogenic components. This was then used as a basis to interpret the IN and CCN data. Instruments and filter types were identical at all sites. Filter samples were collected over 24 hour periods. Instrumental aerosol measurements were made every 12 hours. This provided three measurements for each filter sample. Filter samples were used to determine IN concentrations and aerosol morphology as well as chemistry of the aerosol. Instrumental methods were used to determine the concentrations of CCN and CN (condensation or Aitken nuclei). The data thus provides enough information to determine the daily variability of the aerosol as well as an estimate of the differences between the seasonal mean aerosol (winter vs. summer). Details of the techniques used are described later.

4.4.2 Ojectives

Several elements are used as tracers for the major aerosol sources: natural marine, natural crustal (soil) and anthropogenic. Aluminum is used as the natural crustal aerosol source tracer. The entire mass of aluminum determined in the aerosol particles is assumed to have originated from natural crustal or local soil sources. Based upon this assumption, and knowledge of the ratios of the elements of interest with respect to aluminum in the earth's crust or local soils, the component of an element due to these natural sources can be estimated. This component of an element of interest is then subtracted from the total mass of the element determined in a bulk aerosol collection, and labeled as non-crustal in origin. The calculation used for noncrustal vanadium is given by Equation (4.1).

$$\underset{\text{Vanadium}}{\text{Noncrustal}} = V_{\text{Total aerosol}} - \left[\left[\frac{V}{Al}\right]_{\text{crust (soil)}} \times Al_{\substack{\text{Total}\\ \text{aerosol}}}\right] \qquad (4.1)$$

This calculation can be used to determine the marine sodium present in a given aerosol particle sample as well. All the sodium present in the aerosol particles after subtracting the sodium determined by the assumed sodium/aluminum ratio in the crustal (soil) reference material, is labeled as marine (noncrustal) sodium. Equation (4.2) is used for this calculation.

$$\underset{\text{Sodium}}{\text{Marine}} = Na_{\text{Total aerosol}} - \left[\left[\frac{Na}{Al}\right]_{\text{crust (soil)}} \times Al_{\text{Total aerosol}}\right] \qquad (4.2)$$

The residual sodium is used as a tracer for the marine primary aerosol particle component. Finally, nonmarine sulphate is determined in the

following manner: the product of the marine-sodium concentration calculated above with the known sulphate/sodium ratio in seawater (0.25) results in the concentration of sulphate derived from sea-salt aerosol particles. This component is subtracted from the total bulk aerosol sulphate. The residual is labeled nonmarine sulphate. Equation (4.3) is used for this calculation.

$$\text{Non-marine Sulphate} = SO_4^{=}{}_{\text{Total aerosol}} - \left| \left[\frac{SO_4^{=}}{Na} \right]_{\text{seawater}} \times Na_{\text{Marine aerosol}} \right| \quad (4.3)$$

This residual sulphate is assumed to have originated from a gas-to-particle conversion process, primarily associated with the combustion of fossil fuels. The component of primary marine sulphate in the Arctic is removed by this procedure. The Arctic sites studied are all, with the exception of Poker Flat, in marine environments. Marine aerosol in these locations is an important component of the total aerosol composition. In order to estimate the man-made sulphate fraction, the natural-marine fraction must be accounted for. Since knowledge of natural gas-precursors of sulphate is limited, no estimate of sulphate arising from these sources is made.

Whenever possible, the composition of local soils was used as a reference material from wich the element/aluminum ratios necessary for these calculations were derived instead of a globally averaged crustal rock composition. Local soil and dust were collected and analyzed for their trace element composition by the same analytical procedure as that used for the aerosol particle samples. This was done for the Poker Flat, Pangnirtung, and Ny Alesund-YMER-80 locations. The known

composition of the local rock, a basalt associated with the Mid-Atlantic Ridge of which the island of Iceland is an ocean surface manifestation, was used for the Iceland calculations of non-crustal vanadium. Table 4.2 lists the mass ratios of the elements to aluminum determined in this study. A complete listing was not available for the Icelandic site, Irafoss. The global crustal average as given by Mason (1966) was used to interpret the Barrow, Alaska and Narragansett, Rhode Island aerosol particle composition. For completeness, the elemental ratios relative to aluminum in the crust are also listed.

4.4.3 High-Volume Aerosol Collection

A high-volume aerosol sampler was used at all locations except Pangnirtung, NWT, Canada. A low-volume filter (50 l/min) 47 mm in diameter was used at this site because of logistic and power constraints. At Ny Alesund, Spitsbergen a 14 cm diameter filter was used on the sampler provided by NILU. This sampler provided a flow rate of about 850 l/min. All other sites utilized a sampler having a flow rate of approximately 1700 l/min through a 20 x 25 cm filter. All filters used for chemical analyses were Whatman No. 41 which had undergone an acid-leaching process prior to sampling. This was done to lower the background levels of elements in the filter in order to improve their detection limit. By this method, blank filter concentrations of most elements were reduced by 20-80% of their original values in unwashed filters. This allowed the detection of some elements which would not have been possible otherwise.

4.4.3.1 Non-destructive Neutron Activation analysis (NNAA)

All neutron activation analyses were done at the Rhode Island Nuclear Science Center. The technique permitted the routine analysis of

TABLE 4.2

Elemental composition ratios for local soils relative to aluminum for arctic aerosol particle sampling sites.

Element	Ratio to Aluminum (Element/Aluminum)				
	Poker Flat	Pangnirtung YMER-80	Ny Alesund-Crust	Irafoss	Global Average
Al	1	1	1	1	1
Dy	3.4×10^{-4}	5.6×10^{-4}	2.1×10^{-4}	--	3.7×10^{-5}
Ba	1.5×10^{-2}	3.4×10^{-2}	6.6×10^{-3}	--	5.2×10^{-3}
Ti	4.9×10^{-2}	0.11	5.7×10^{-2}	--	5.4×10^{-2}
I	$<1.4 \times 10^{-4}$	7.9×10^{-4}	$<2.8 \times 10^{-4}$	--	6.1×10^{-6}
Br	$<8.8 \times 10^{-7}$	2.0×10^{-3}	2.8×10^{-4}	--	3.1×10^{-5}
Cl	1.6×10^{-2}	0.40	1.1×10^{-2}	--	1.6×10^{-3}
Mn	1.6×10^{-2}	1.8×10^{-2}	4.5×10^{-3}	1.9×10^{-2}	1.2×10^{-2}
Mg	0.96	0.50	0.15	--	0.26
V	1.4×10^{-3}	1.9×10^{-3}	1.9×10^{-3}	4.2×10^{-3}	1.7×10^{-3}
Na	0.20	0.67	0.13	--	0.35
Ca	0.17	0.20	0.12	--	0.45
In	$<3.1 \times 10^{-6}$	3.0×10^{-6}	$<8.2 \times 10^{-7}$	--	1.2×10^{-6}

* From Mason, 1966

fifteen elements in the aerosol. The analytical scheme used for the irradiation and counting of the aerosol samples was developed by Dr. Kenneth A. Rahn at the Center for Atmospheric Chemistry Studies for analysis of arctic aerosol samples. The irradiations were performed in the 2-MW nuclear research reactor located on the grounds of the Graduate School of Oceanography at the University of Rhode Island. Approximately 15 cm^2 of filter were irradiated representing 60-80 m^3 of air sampled. Gamma-ray spectra of the irradiated samples were acquired using an Ortec 38 cm^3 Ge(li) gamma-ray detector (7% efficiency) coupled to an Ortec Model 440 amplifier and a Nuclear Data Model 2200 4096-channel analyzer. Spectra were written on magnetic tape by an Ampex Model 7211 7-track recorder and then processed by computer.

Each filter sample was irradiated for five minutes at a thermal neutron flux of 4×10^{12} n cm^{-2} sec^{-1}, cooled for two minutes, and counted for 400 seconds clock-time. After fifteen minutes of total cooling time the sample was counted again for 1000 seconds clock-time. A neutron flux monitor was also irradiated with each sample and counted for 80 seconds clock-time after a cooling period of twelve minutes. This permitted corrections to be made in the final analysis for any fluctuations in the neutron flux during the irradiation. A pulser was used to correct for dead-time losses during counting. Standard solutions, consisting of known amounts of the elements spotted onto the same type of filter paper, were run according to the same procedure and using the same flux monitor. Blank filters, which underwent the same handling process as a sampled filter, were analyzed by the same procedure. Blank filters were analyzed for each sampling site and period. These analyses permitted the determination of up to 15 elements

resulting from the irradiation includings: ^{52}V, ^{28}Al, ^{24}Na, ^{128}I, ^{56}Mn, ^{38}Cl, ^{80}Br, ^{27}Mg, ^{66}Cu, ^{51}Ti, ^{125m}Sn ^{49}Ca ^{116m}In, ^{139}Ba, and ^{165}Dy,

4.4.3.2 Sulphate analysis

In addition to the elemental analysis by NNAA, all the high volume aerosol samples were analyzed for their water soluble sulphate content. These analyses were done by two different methods. All samples except those from the Ymer expedition were analyzed for sulphate using a turbidimetric technique. Aliquots of the filter samples were placed in distilled de-ionized water for a period of 24 hours to leach the sulphate. A barium sulphate precipitate was then formed and kept in a colloidal suspension using a gelatin reagent. This suspension was then placed in a 5 cm cuvette. Analyses were performed using a Varian Series 634 Spectrophotometer at a wavelength of 420 nm and slit width of 1.0 nm. Standard sulphate solutions of known concentration were also run by the same procedure before and after each series of determinations. The procedure is based upon the one described by Tabatabai (1974).

The samples from the Ymer expedition were analyzed using a Dionex 16 Ion Chromatograph. Standard sulphate solutions were run before and after each series of samples analyzed on the ion chromatograph. Nine parts of the sample sulphate solution were mixed with one part of the eluent consisting of a 0.003M $NaCO_3$ + 0.0024M Na_2CO_3 solution. The comparison of standards with samples was done using peak height analysis of the conductivity output from the ion chromatograph.

4.4.4 Low-Volume Aerosol Collection

Filter samples were collected on 0.2μm pore-size Nuclepore filters for 24-hour periods at all sampling locations. These samples were later analyzed to determine IN concentrations and aerosol-particle morphology.

4.4.4.1 Ice nucleus concentration

Ice-nucleus concentrations were determined from particles collected on 0.2-μm pore size, 37 mm diameter hydrophobic Nuclepore filters, using a modified static thermal gradient diffusion chamber (STGDC) and a technique similar to that described by Stevenson (1963). Nuclepore filters were chosen because of their well-documented filtering characteristics, excellent thermal conductivity as a result of their small thickness and ease of mounting in the STGDC. Briefly, the filters were mounted in Vaseline-coated trays (two to a tray, four trays) and placed in the STGDC. The distance between the ice surface and filter surface was 0.5 cm. The filters were maintained at temperatures of -16^{o}, -20^{o}, -24^{o}, and -28^{o}C while the ice surface was maintained at a temperature which provided a supersaturation of 1% with respect to a plane water surface. The filters were kept in the STGDC for a 45-minute period, then the number of ice crystals grown on the filter surface were counted. A blank filter was analyzed with each run, and a blank value for each set of samples in a run was determined by taking the average of three runs centered on the one of interest.

Each IN collection in the field utilized of a set of four filters. One was kept covered and served as a blank and was exposed to the same handling procedures as the other sample filters at each site. A second filter was designated a "high volume" IN filter. A flow rate was maintained through this filter to provide a sample volume of approximately 1000 liters over a 24-hour period. The remaining two filters were designated "low volume" IN filters and were sampled at flow rates which provided sampled volumes of 100-300 liters over a 24 hour period. These procedures were undertaken for two reasons: 1) The

low overall volumes were used because of the known vapor depletion effect of too many ice crystals growing per unit area on a filter surface and the presence of CCN in the total aerosol collected also competing for the available water vapor, and 2) it was expected that there would be greater concentrations of IN at colder analysis temperatures. Thus the "low volume" filters were used for the colder temperatures and the "high volume" filters were used for the warmer temperatures. In addition, all IN concentrations reported reflect the correction factor as discussed by Huffman and Vali (1973). The factor is based upon the number of ice crystals observed per unit area on the filter and the concentrations of CCN active at 1.0% supersaturation as determined by the CCN counter used during the filter sampling.

4.4.4.2 Scanning electron microphotographs

A 47-mm-diameter Nuclepore filter was used to sample the aerosol for later study using a scanning electron microscope. The filter sample represented the aerosol for a 24 hour period, collected concurrently with the high and low volume filter samples, at a flow rate of approximately 40 lpm. Thus about 50-60 m^3 was sampled. The samples were collected to aid in the interpretation of the chemical composition of the bulk aerosol and the cloud-active aerosol observations. Information on aerosol morphology, numbers and sizes were obtained from these samples.

4.4.5 Instrumental Aerosol Particle Data Collection

In addition to filter samples, the aerosol was also monitored using instrumental techniques in the field. Two different instruments were used for routine measurements.

4.4.5.1 Static-thermal-gradient diffusion cloud chamber

A thermal-gradient-diffusion chamber was used to determine the concentrations of CCN at 0.2%, 0.5%, 1.0% and 2% supersaturation with respect to water. The instrument used was a Mee Model 130 Cloud Condensation Nucleus Counter (Serial #3). The original optical bench, light source, and detection components were removed and replaced with a simple laser (He-Ne)-illumination-photographic counting system. This permitted discrete droplet counting from the photographic film. This extended the minimum detectable concentration down to 10 cm^{-3}. Details of the instrument and its performance are given in Table 4.3 See also Borys (1981), Appendix D.

4.4.5.2 Gardner small particle counter

To determine the concentration of Aitken or condensation nuclei ($r < 0.1$ μm) a Gardner Small Particle Counter was used. This counter is a rapid expansion type, developing supersaturations greater than 200%. The results from such a counter are a measure of the total number concentration of the aerosol which is dominated by the aerosol in the Aitken size range. The counter was typically used as a local contamination detector since it is very sensitive to fresh aerosol particles from nearby combustion sources. In addition to the regular measurements made twice a day concurrently with the CCN counter, measurements were made whenever the sampling site was visited or the potential for local contamination arose. In this way, the filter samples collected were assured to be representative of the region and not of the locality. If a strong source of local contamination was detected, the main filter sampling was discontinued until the proper conditions returned.

TABLE 4.3

CCN Counter Specifications

Detection System:	
Laser light source	5 milliwatts 0.65 micron wavelength
Photographic counting	2475 recording film ASA 3000
Minimum detection size	$\approx$ 1 μm radius
Sampling:	
Flow rate	4 lpm for 10 sec
Sample volume	0.011 cm^3
Time for spectra (4 pts)	30-90 min
Chamber Specifications:	
Aspect ratio	4:6:1
Temperature control	Thermoelectric cooling of lower plate
Usable supersaturation range	0.2 - 2.0%

4.5 Meteorological Analysis

Initially, isentropic or isobaric airmass trajectory analyses were to be determined for each 24-hour filter sample to aid in the interpretation of the aerosol chemistry and to provide information on aerosol source regions. A study of the validity of these analyses in the Arctic was undertaken to determine whether the information they would provide would be useful in the context of this work. The results of this study are presented in Appendix B. Trajectory analyses, although providing what appears to be a definitive history of an air parcel actually are only as good as the data (upper winds) used to construct them. Since the data over the Arctic are sparse and heavily interpolated, results from trajectory analysis would be questionable.

Still, information on the meteorology, especially the wind regime existing during collection of the samples, is necessary. A simple representation of the winds is a multilevel-isohypse analysis. This is a depiction of a constant pressure surface contour line, extending upwind from the sampling site, and parallel to the analyzed contour lines from the maps of the National Meteorological Center (NMC). The wind speed is estimated from contour spacing for a level over the site, and the isohypse is drawn a length representative of the distance an air parcel would have traveled in 3 days using this wind speed. Thus the isohypse analysis depicts the important wind direction and the relative speed. This analysis is quick and simple, and probably provides as much information as an isobaric trajectory analysis in the Arctic.

Isentropic trajectory analyses, which normally would give a more detailed and accurate depiction of the air mass history in three dimensions, would be more in error and less reliable than the isobaric

trajectory. This would result from the need to determine a conservative property of the atmosphere as well as the winds. Additional uncertainty would be introduced in the final result since the conservative property would also have to be an interpolated parameter.

Based upon these arguments, the isohypse analysis is best suited for the kind of meteorological analysis and atmospheric transport information necessary for this study. Isohypse analyses were performed for each sample collected at each site. The isohypses are plotted and presented in the detailed field experiment discussions for each site in Appendices E through J.

V. RESULTS

The measurements of the cloud-active aerosol and trace-element chemistry in the Arctic were designed to determine the importance of long-range transport of aerosol particles from mid-latitudes on the total Arctic cloud-active aerosol concentrations. Summer and winter experiments have been completed at each location described previously. The data from each site have been summarized in the form of plots, tables and elementary statistical analyses. Isohypse analyses of the 850 mb and 700 mb pressure surface weather maps have been completed for each site study-period. In addition, scanning electron photomicrographs of the aerosol were taken to assist in the interpretation of the other aerosol data. These are used together in discussions of the results from each site study presented in Appendices E through I. In this chapter, an overview of the results of the study on arctic cloud-active aerosol will be presented. For more detailed analysis and interpretation for each location, refer to appendices E through J.

5.1 Representativeness of the Study

The field experiments were conducted over short (7-11 day) periods. As a first step it is necessary to know the representativeness of the results from these experiments in a climatological sense. The meteorological conditions at the time of study for the different locations and the time of the year were compared to the conditions over a long-term prior to the experiment. As a first estimate the mean atmospheric temperature and pressure were calculated for each arctic

field experiment period. These means were then compared to the long-term mean temperature and pressure for the same days of the year for 5 to 7 years prior to the experiments. Climatological data were used from the years 1972 to 1978. The nearest regularly reporting upper air station was used for each site.

Table 5.1 summarizes the short-term (field experiment) and long-term (climatological) mean temperature and pressure for the arctic sites. In all cases except for atmospheric pressure during winter at Poker Flat and Pangnirtung, the mean value for the temperature and pressure fell within one standard deviation of the long-term mean. The last column shows the results of the student's 't' test used to compare the short- and long-term means. The value of P is the probability that the difference between the climatological and site visit means of temperature and pressure for a given site and season is not due to chance. Values of $P < 0.01$ indicate a significant difference may be present. These occur for the sites of Poker Flat, Pangnirtung and Irafoss. There is no indication of significant differences between the long- and short-term temperature and pressure for the high arctic sites of Barrow and YMER-80/Ny Alesund. None of the sites had values of P less than one for both temperature and pressure. Combining this with the observation that a P value < 0.01, and a short-term mean value of pressure or temperature exceeding one standard deviation from the long-term mean occurred only once (for Poker Flat, winter, pressure) shows that the overall data set was collected during periods of typical meteorological conditions. The interpretation of the aerosol data from these sites as being regionally representative is therefore appropriate.

TABLE 5.1

A Comparison of the Mean Temperature (°C) and Pressure (mb) during the Arctic Field Site Visits to a Long-Term Mean Temperature and Pressure for the Same Periods.

Site	Season		Site Visit Mean	Long-Term Mean		P*
Barrow	Summer	T	6.5±3.9	5.7±4.8	(6 yr)	0.42
		P	1014.6±2.3	1014.1±5.7		0.50
	Winter	T	-23.3±4.0	-24.8±5.6	(7 yr)	0.21
		P	1022.8±2.9	1021.1±8.2		0.35
Poker Flat	Summer	T	16.1±5.3	16.8±5.3	(6 yr)	0.50
		P	1012.2±5.2	1004.9±11.0		0.02
	Winter	T	-0.6±5.3	-1.9±6.8	(7 yr)	0.30
		P	989.6±5.6	1004.3±11.9		<0.01
Pangnirtung	Summer	T	3.8±1.7	6.2±3.5	(6 yr)	< 0.01
		P	1009.3±8.2	1007.6±6.3		0.08
	Winter	T	-19.6±6.2	-27.3±10.0	(6 yr)	< 0.01
		P	1013.4±9.6	1004.2±8.6		0.02
Irafoss	Summer	T	10.1±2.2	6.7±5.2	(7 yr)	< 0.01
		P	1008.6±7.9	1007.8±8.5		> 0.50
	Winter	T	4.0±2.1	1.1±3.4	(7 yr)	< 0.01
		P	993.9±7.6	997.8±14.9		0.34
YMER-80	Summer	T	4.5±2.5	4.2±1.9	(5 yr)	0.31
		P	1013.2±5.0	1011.1±8.5		0.11
Ny Alesund	Winter	T	-16.1±4.5	-12.2±9.5	(7 yr)	> 0.50
		P	1009.1±7.0	1008.8±12.5		0.20

P* is the probability that the difference between either the mean temperature or pressure values for a given site and season is due to chance. Values of P < 0.01 indicate the two means are significantly different based upon the Student's t Test.

5.2 Arctic Cloud Condensation Nuclei

The measurements of cloud-condensation nuclei at all the arctic locations were routine and sensitive enough for the lowest concentration encountered (see the description of the CCN counter in Appendix D and Chapter IV). Discussions of the results of the CCN measurements will deal primarily with the concentrations determined at 0.2% supersaturation. There are four reasons for this. 1) This investigation is directed at the nucleational aspects of Arctic Haze aerosol. Haze particles form at low supersaturations with respect to liquid water in the atmosphere. The CCN counter does not measure actual haze nuclei which are generally assumed to be unactivated CCN at very low supersaturations (<0.1%). The lowest supersaturation the CCN counter can be operated at with precision is 0.2%. This supersaturation was chosen to be studied in detail so as to lie nearest the haze forming range of supersaturation. 2) The CCN at this supersaturation were found to vary over a broader range of concentrations than at any other supersaturation. This suggests that these nuclei may be more sensitive for use in determining causes for CCN variation as a result of changes in the total aerosol present. The concentrations of CCN at these supersaturations were always varying in the same sense as at the other supersaturations so that conclusions drawn apply to the entire CCN spectrum. 3) It was observed that the CCN active at 0.2% supersaturation were not effected by local fresh aerosol sources as a result of man's activities. This eliminates possible local contamination, and suggests 0.2% supersaturation may be most sensitive to an aged aerosol, transported over a long distance, and more representative of the region. 4) Finally, the dominant cloud form in

the Arctic is stratiform. These clouds do not form as a result of large supersaturations in the atmosphere. The CCN measurements at 0.2% should best represent the nuclei most likely to play a role in arctic cloud formation.

Results of the mean CCN concentrations for summer and winter at 0.2% supersaturation are presented in Table 5.2 for all the field locations. The last two columns are the winter/summer ratios of the mean concentrations of the pollutant aerosol tracers, noncrustal vanadium and nonmarine sulphate. A similar winter/summer ratio for the CCN is given in the center column. These comparisons were made because of the strong seasonal pollutant aerosol variation in the Arctic. Any dependence of the CCN concentrations to the pollutant aerosol concentrations should be evident in this comparison.

The two high-arctic sites, Barrow and Ny Alesund-YMER, provide the best examples representative of the true polar arctic airmass. It is evident that the winter/summer ratio of both the CCN and the pollutant tracers are significantly greater than one. The winter/summer ratios of the other natural aerosol tracers used in this study are an order of magnitude smaller than the ratios of the pollutant tracers. The presence of the pollutants during the winter are associated with higher CCN concentrations, approaching a factor of ten at Ny Alesund-YMER.

Table 5.3 presents the percent variance of both the IN and CCN concentrations explained by the different aerosol tracers used. The variance values are the square of the linear correlation coefficients determined between the nuclei and the aerosol tracers. Values significant at greater than 95% are indicated by underlining. Those in parentheses additionally showed homogeneity of variance at the same

TABLE 5.2

SEASONAL MEAN CONCENTRATION OF POLLUTANT AEROSOL TRACER COMPONENT AND CLOUD CONDENSATION NUCLEI AT .2% SUPERSATURATION

Sampling Location	CCN Concentration (CCN cm^{-3})		Winter/Summer Ratio	Aerosol Winter/Summer Ratio	
	Winter	Summer		Noncrustal Vanadium	Nonmarine Sulphate
Barrow	150	35	4.3	>45	>38
Ny Alesund-Ymer	240	30	8.0	11	29
Poker Flat	100	170	0.59	2.5	8.3
Pangnirtung	60	25	2.4	7.7	---
Irafoss	105	110	0.95	0.08	<0.40
Narragansett	500	2760	0.18	2.0	1.0

TABLE 5.3

PERCENT OF VARIANCE EXPLAINED OF ICE NUCLEI AT -20° AND 1% SUPERSATURATION AND CLOUD CONDENSATION NUCLEI AT 0.2% SUPERSATURATION BY AEROSOL SOURCE TRACERS AT ARCTIC AND SOURCE REGION SITES.

SITE	SEASON	CLOUD ACTIVE AEPOSOL	AEROSOL SOURCE TRACERS NONCRUSTAL Al	Na	I	NONCRUSTAL V	$SO_4^{=}$	NONMARINE CN
Barrow	winter	IN	-17	-24	12	(68)	15	40
		CCN	-6	-4	6	68	75	55
	summer	IN	-1	(56)	-23	-9	6	3
		CCN	-33	20	-13	-14	-4	-38
NY Alesund	winter	IN	-2	-1	1	-5	-4	-9
		CCN	10	1	(82)	(78)	(93(	(98)
YMER-80	summer	IN	-1	-1	-4	0	(93)	79
		CCN	19	20	14	30	57	24
Irafoss	winter	IN	34	16	31	25	12	42
		CCN	49	(56)	(57)	15	-7	(73)
	summer	IN	22	-7	0	-5	0	0
		CCN	(48)	32	(73)	57	(75)	0
Poker Flat	winter	IN	17	17	-9	(-48)	27	35
		CCN	-47	-11	(52)	8	1	-35
	summer	IN	-15	-6	-2	0	-7	15
		CCN	4	42	40	17	44	-2
Pangnirtung	winter	IN	13	0	1	-25	10	-6
		CCN	43	2	-1	-9	-4	-16
	summer	IN	69	-8	6	0	---	41
		CCN	-10	-1	16	0	---	0
Narragansett	winter	IN	-6	32	39	(75)	-13	11
		CCN	2	-17	-4	-2	31	-12
	summer	IN	48	0	25	66	39	47
		CCN	(94)	(57)	(82)	(88)	(79)	6

Underlined values denote correlations for small sample sets significant at the 95% level, negative values denote negative correlations, parentheses indicated the significantly correlated sample pairs with homogeneity of variance significant at the 95% level.

level of significance. These were all derived from the time-series data gathered at each site.

Concentrating on the Barrow and Ny Alesund-YMER data, it is evident that the CCN variability from day to day is also closely related to the variability of the pollutant tracers. This close association occurs during the winter, when the pollutants are present, and disappears in the summer, when they are absent. As discussed in Appendix J, the large summer value for the CCN versus sulphate variance during the YMER expedition is due to only three of eleven samples which were collected while the ship was south of the polar front. This is further evidence that the pollutant aerosol plays an important role in both the concentration and day-to-day variability of the CCN in the Arctic.

The lower-latitude arctic sites of Poker Flat, Pangnirtung and Irafoss do not present the same clear picture. The reasons for this are discussed in detail in the appropriate appendices. The primary reason for the difference is due to the different climatology of these sites. Poker Flat is essentially an interior continental location, and is separated from the arctic polar airmass by the Brooks Range in Northern Alaska. Airmasses reaching this location are more likely to arrive from the North Pacific. The same is true of both Pangnirtuang and Irafoss which are strongly influenced by North Atlantic rather than Arctic airmasses. Both of these maritime airmass source regions are relatively pollutant free and have a maritime aerosol.

The data from Poker Flat and Irafoss reflect the maritime influence. Table 5.3 shows the CCN were more closely related to the maritime aerosol tracers, marine sodium and iodine, at these sites. Pangnirtung is the most remote site studied based upon the very low

overall aerosol concentrations measured there. As a result, the aerosol data had a great deal of scatter and, as can be seen in Table 5.3, no significant correlations could be detected between the CCN and the aerosol tracers. The evidence in Table 5.2 that winter had elevated pollutant aerosol and CCN concentrations relative to summer was due to a small increase in absolute concentration resulting in a relatively large increase in the winter/summer ratio. Table 5.2 shows elevated pollutant concentrations were detected at Poker Flat in winter, but CCN concentrations were actually lower. Although some transport of arctic air can reach Poker Flat during winter, the continentality of the region and transport of maritime air masses from the south during summer provide more sources of CCN. Poker Flat is not a true arctic site based upon the measurements made during this study. The concentrations of noncrustal vanadium during the winter experiment were ten times lower than those found a Barrow. Intrusions of polluted arctic airmasses to Poker Flat are sporadic and not routine.

Irafoss is located well outside of the true polar arctic and lies along the North Atlantic storm track. As mentioned previously, maritime aerosol sources are strong. Although there is little contrast between the mean winter and summer CCN concentrations, there is a large difference in the concentrations of pollutant aerosol (see Table 5.2). In contrast to the other sites, Irafoss had a summer pollutant maximum due to a transport event from Europe (see Appendix I). This event was significant, and its influence on the CCN concentration can be seen in Table 5.3 where the largest variance values for CCN during the summer was associated with nonmarine sulphate. Although not related to the Arctic Haze phenomenon in the north during winter, this is further

evidence of the influence of long-range pollutant transport on CCN concentrations.

Figure 5.1 is a summary plot of the Arctic sites, and the mid-latitude source region site, mean CCN spectra for the winter experiments. Figure 5.2 is a similar plot of the mean CCN spectra for summer. As expected, the source region site of Narragansett has the highest overall CCN concentrations during both seasons. There is a much smaller difference between the CCN concentrations at the source region site of Narragansett and the Arctic sites during the winter. This results from a simultaneous decrease in the CCN concentrations at Narragansett and an increase at most of the arctic sites during the winter season. The CCN concentrations at Ny Alesund (79^{o}N) during winter are only a factor of two less than those at Narragansett. Similarly, the mean nonmarine sulphate concentration at Ny Alesund (5.0 $\mu gm\ cm^{-3}$) is about 65% of the nonmarine sulphate concentration at Narragansett (7.7 $\mu gm\ cm^{-3}$). Ranking the remaining arctic sites by their mean-winter nonmarine sulphate and noncrustal vanadium concentrations produces the same ranking shown for the CCN concentration at 0.2% supersaturation in Figure 5.1. Combining this observation with the results shown in Table 5.3 produces strong evidence that CCN concentrations in the arctic are related to pollutant aerosol tracers in the atmosphere. Applying the same ranking test for all the sites using the other natural aerosol tracers does not produce a similar ranking. Rather, the ranking is different for each of the tracers, with the exception of Narragansett, which has the highest concentrations for all the tracers, reflecting the overall high aerosol loading within the source region.

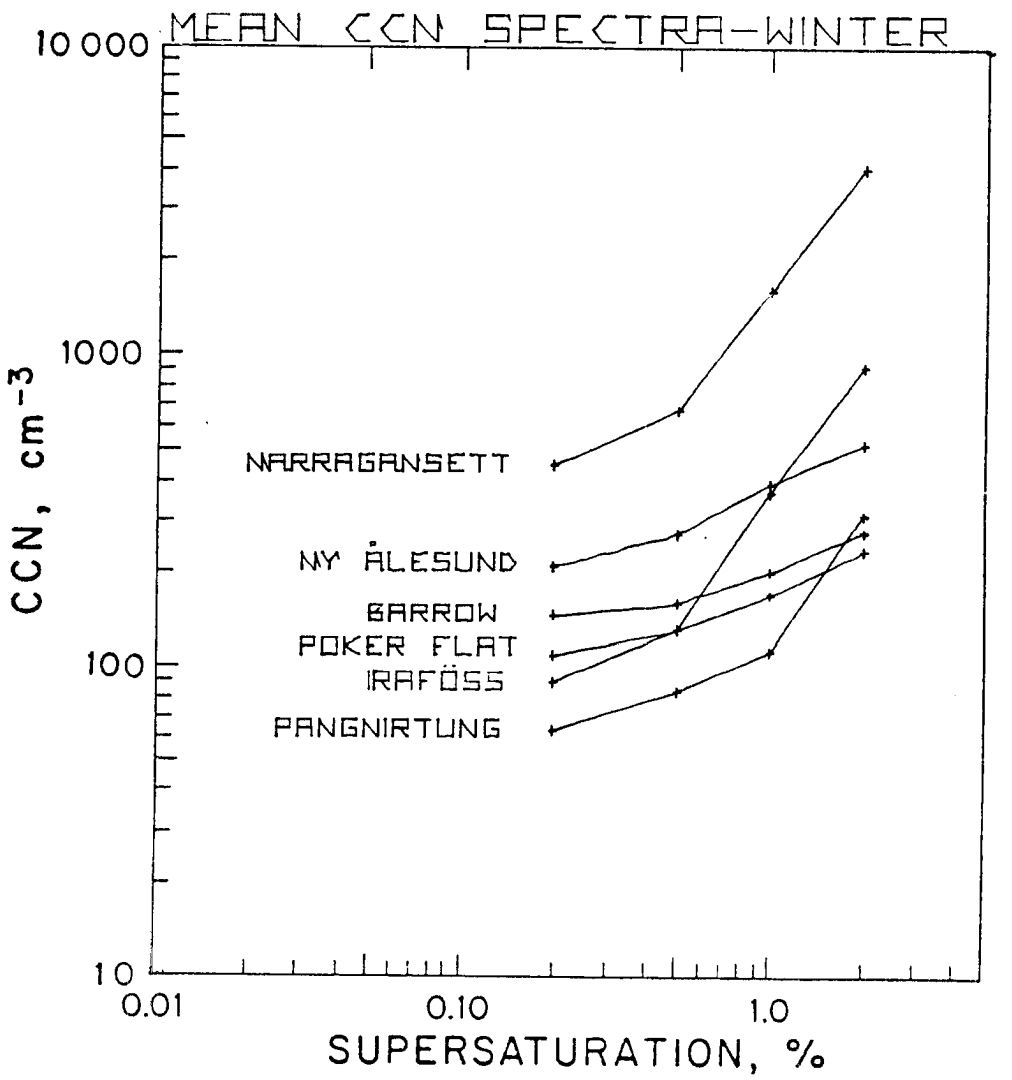


Fig. 5.1 Summary plot of the mean winter CCN spectra for the five arctic aerosol experiment sites and the one mid-latitude source region site.

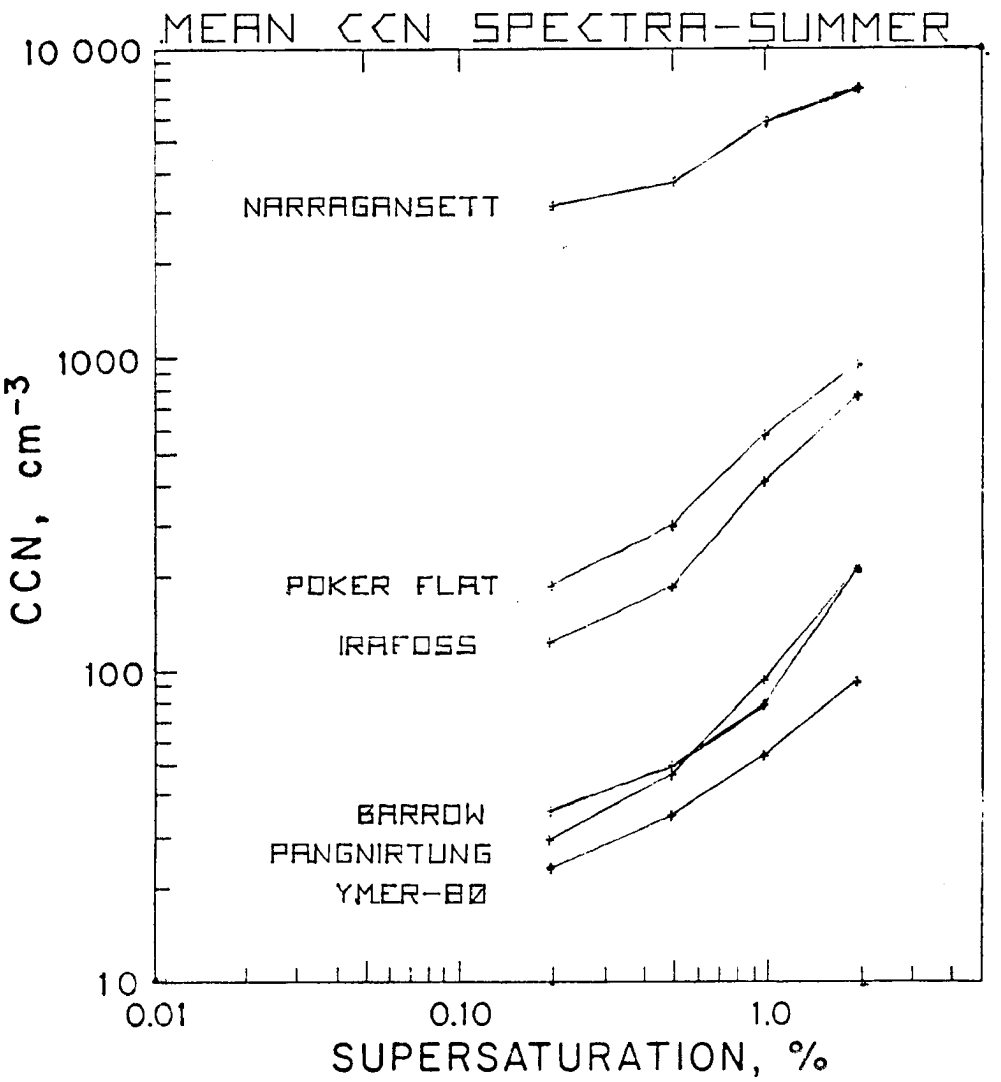


Fig. 5.2 Summary plot of the mean summer CCN spectra for the five arctic aerosol experiment sites and the one mid-latitude source region site.

Scatter-diagram plots of the entire set of data for CCN concentration at 0.2% supersaturation versus the concentrations of the aerosol tracers used reflect the same strong dependence on the CCN concentrations on the pollutant aerosol tracers. Figure 5.3–5.7 are scatter diagram plots of all the data from all five arctic sites for both winter and summer experiments. Crosses are used for each data pair. Dots indicate data where one of the two variables was not detectable due to low concentrations. The dashed line is the best fit linear regression line of the log of the concentrations. Figures 5.3 and 5.4 show the expected positive slope of the regression line for the CCN versus nonmarine sulphate and noncrustal vanadium data. The correlation coefficients from these data are +0.57 between CCN and nonmarine sulphate and +0.65 between CCN and noncrustal vanadium data, both of which are significant at greater than the 99.9% level. Although these correlations are significant, the percent variance explained of the CCN by the noncrustal vanadium and nonmarine sulphate, respectively, is 42% and 32%. These variance values are not as high as those for some of the individual arctic sites (see Table 5.3). Pollutant aerosol tracers can account for a large percentage of the total variance when averaged over all the arctic aerosol data presented.

Figure 5.5 shows the somewhat surprising result of a positive relationship between aluminum and CCN concentrations for the entire data set. The correlation coefficient is +0.34 and significant at greater than the 99% level. There are two possible explanations for this observation which cannot be resolved with the available data. continental areas are known to have higher concentrations of CCN, and aluminum is used here an an indicator of continental airmasses. In

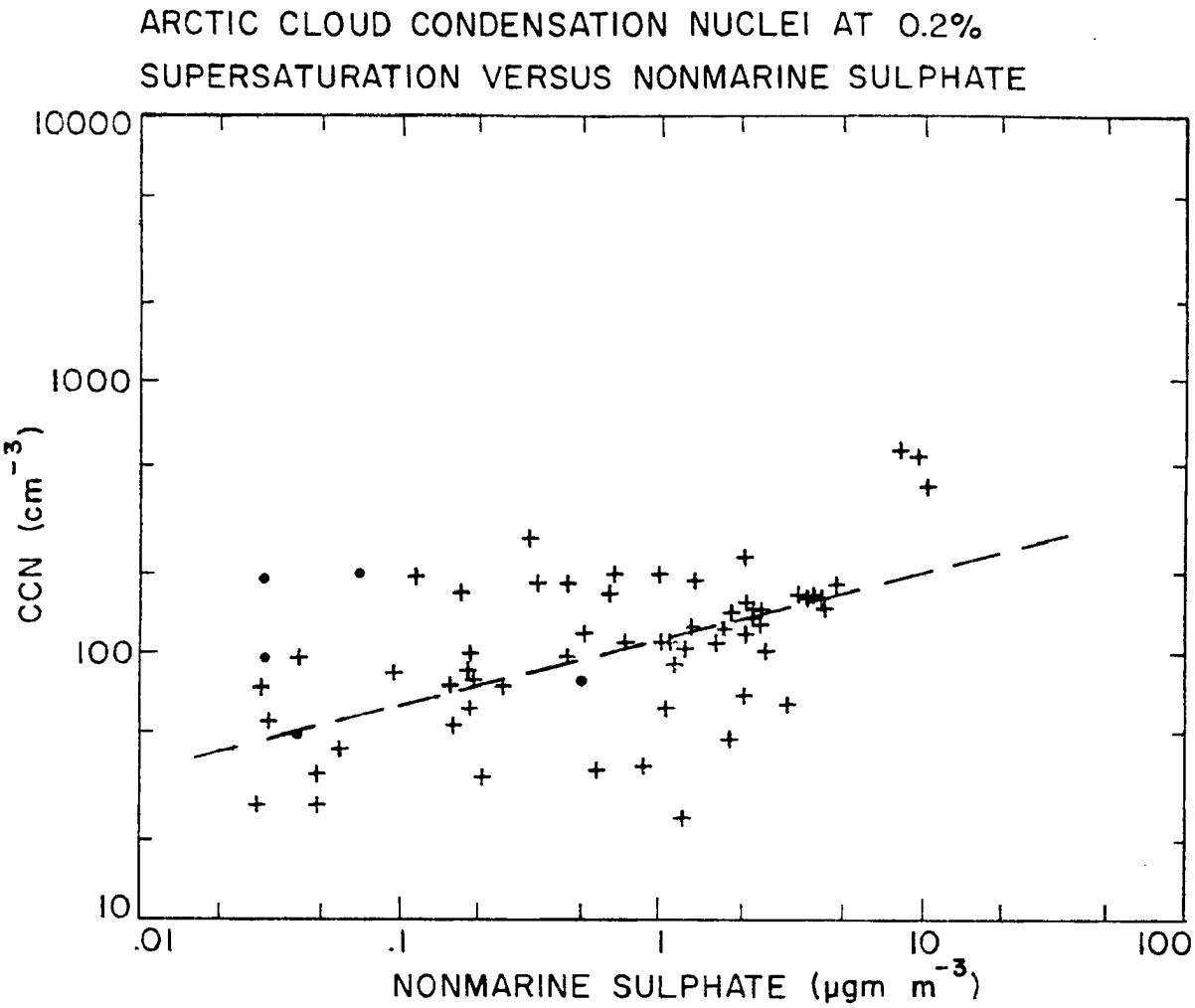


Fig. 5.3 Scatter diagram plot of CCN concentrations vs. nonmarine sulphate aerosol concentrations from all five arctic field site experiments. Crosses indicate data pair values. Dots indicate one or both of the values plotted is a less-than value. Dashed line is the best fit linear regression of the logarithms of the data.

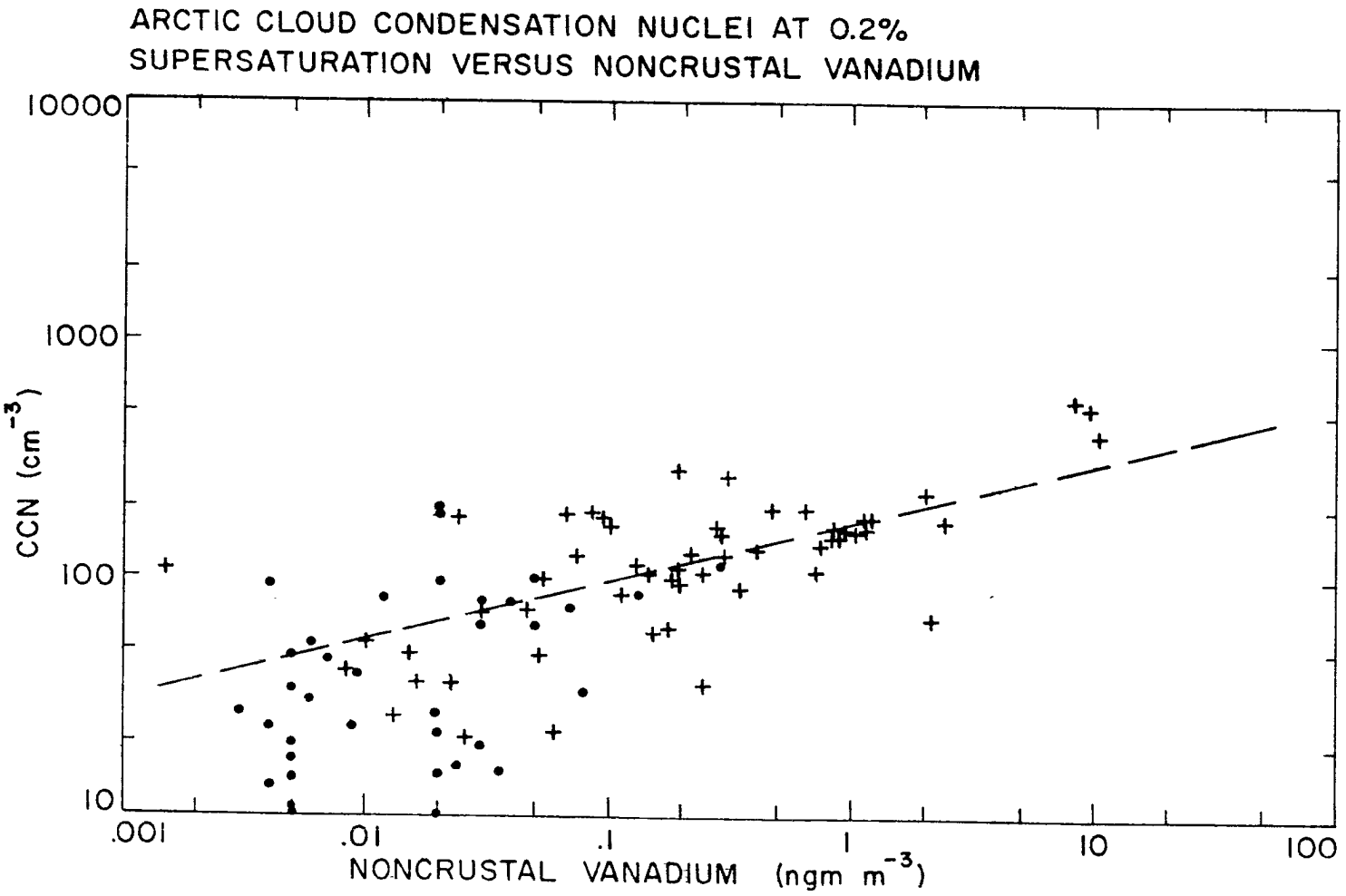


Fig. 5.4 Scatter diagram plot of CCN concentrations vs. noncrustal vanadium----same as Fig. 5.3.

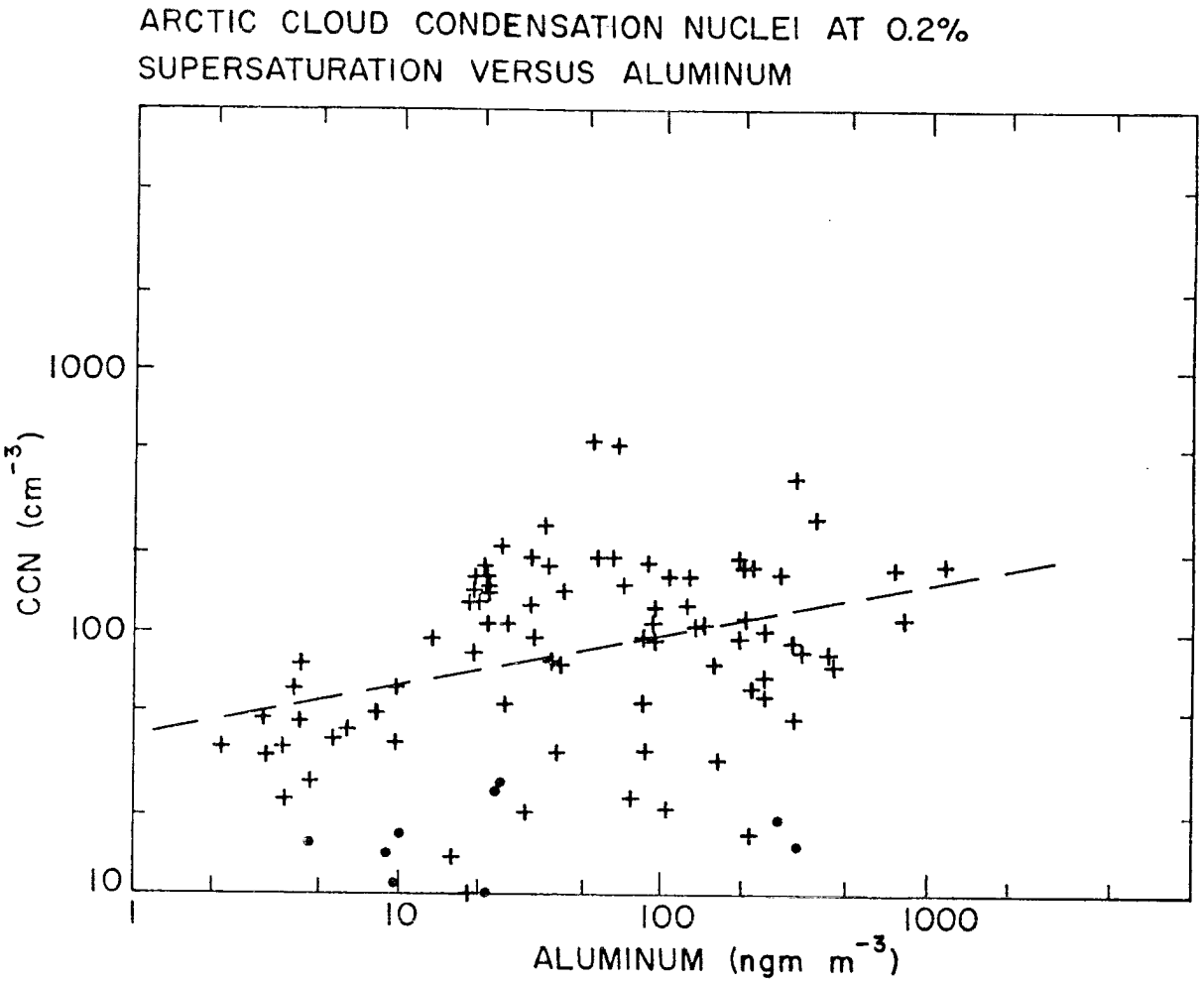


Fig. 5.5 Scatter diagram plot of CCN concentrations vs. total aluminum----same as Fig. 5.3.

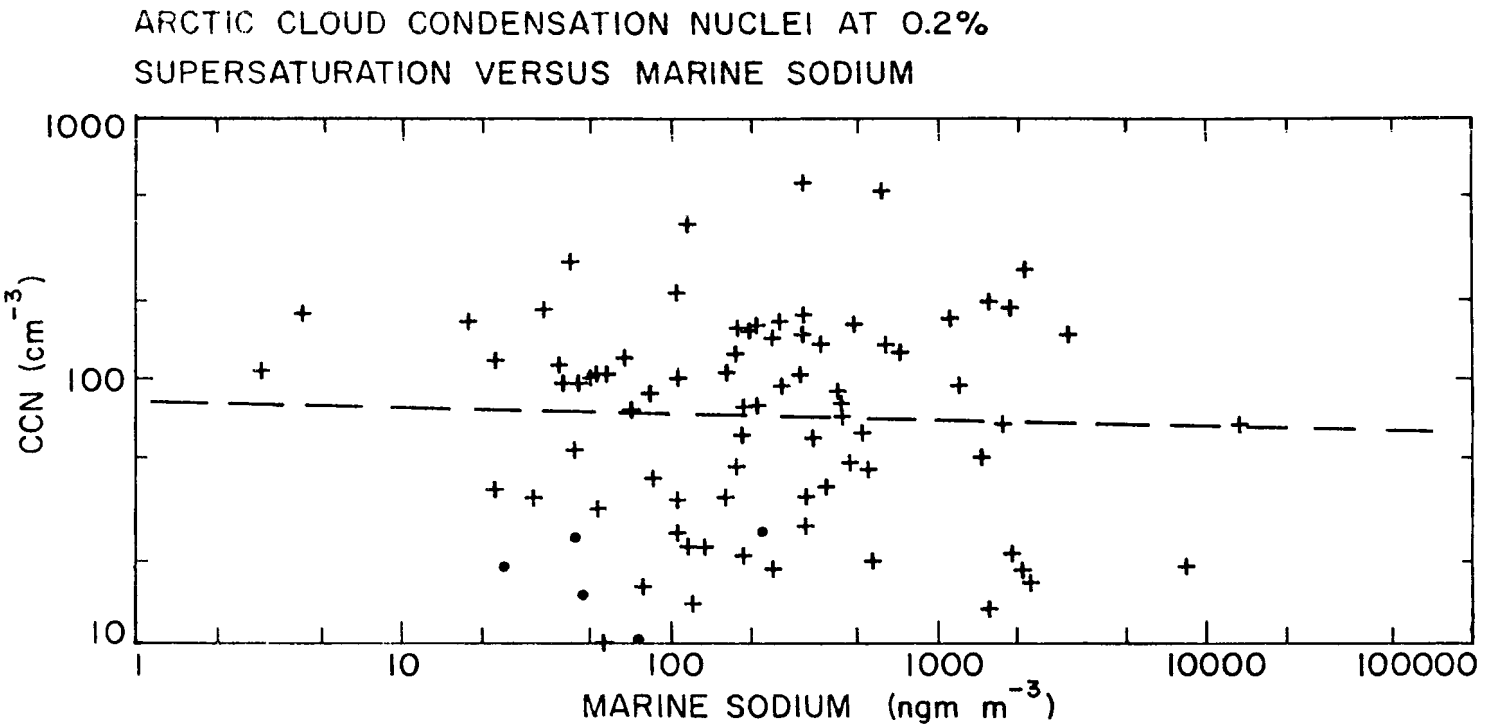


Fig. 5.6 Scatter diagram plot of CCN concentrations vs. marine sodium----same as Fig. 5.3

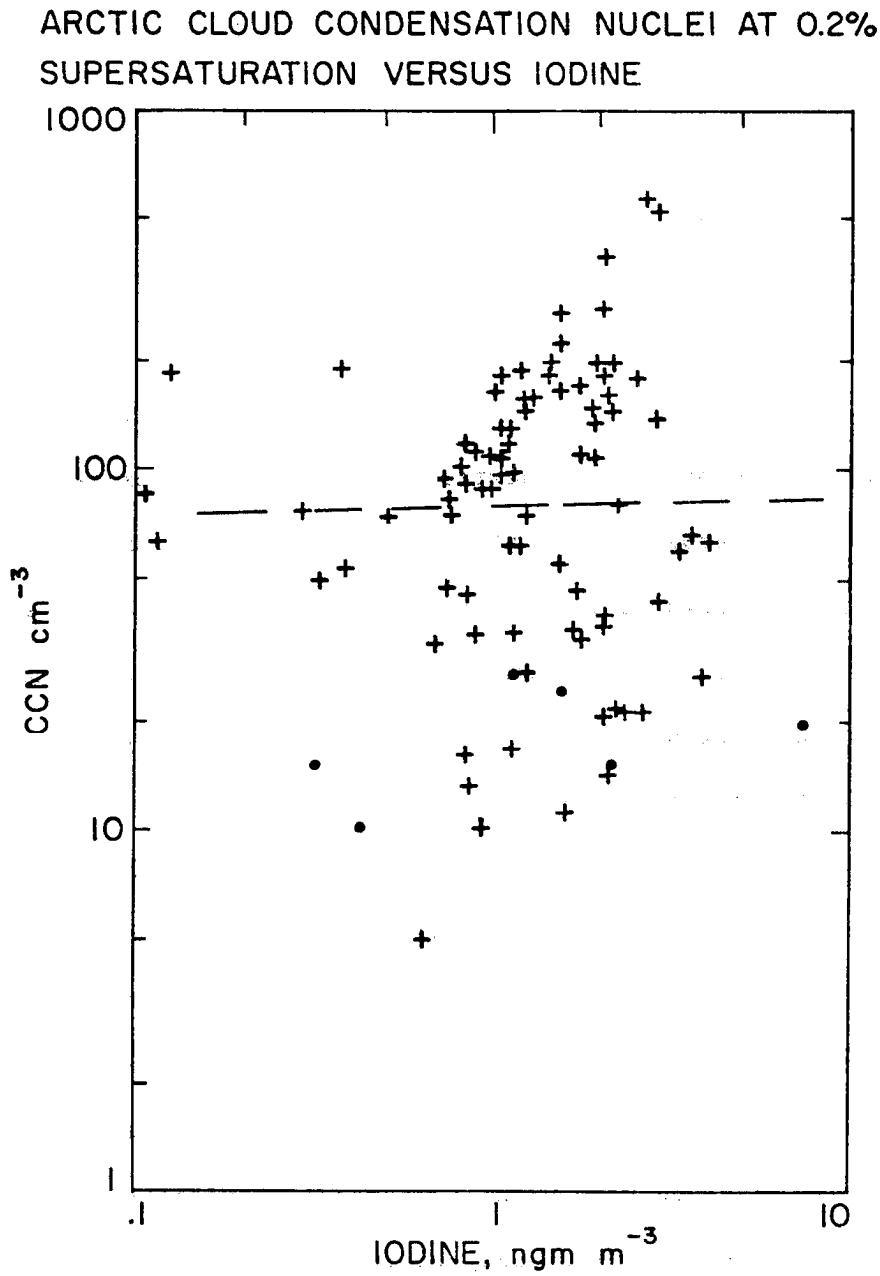


Fig. 5.7 Scatter diagrams plot of CCN concentrations vs. iodine --- same as Fig. 5.3.

addition, pollution sources are located on the continents and to some extent produce aluminum containing particles which could be transported long distances along with the other pollutant aerosol tracers nonmarine sulphate and noncrustal vanadium. The two effects cannot be separated, but are in-line with the observations.

Figures 5.6 and 5.7 show no relationship between CCN and the marine primary and secondary aerosol tracer marine sodium and iodine. This is surprising since the sites are all, with the exception of Poker Flat, located in marine environments. This leads to questions of the importance of seasalt particles acting as the primary source of CCN even in marine areas. This also provides evidence that on a large-scale, secondary aerosol formation, in particular oxidation and hydrolysis of sulphur gases to form sulphate-containing particles, may be the single largest source of CCN in the remote northern latitudes.

5.3 Arctic Ice Nuclei

As in the case of the arctic CCN data, much of the discussion of the result of the IN data will center around the concentrations at one activation temperature or ice supersaturation value. The IN concentrations at $-20^{o}C$ and 1% supersaturation with respect to water (22.6% supersaturation with respect to ice) were chosen for the comparative analyses with the aerosol chemical tracers. The reason for this choice was the quality of the data at this temperature and supersaturation. Very low concentrations, were often encountered at the warmer temperature of $-16^{o}C$. At colder temperatures, errors in the data were introduced by difficulties in the analytical method. The method, described in Chapter IV, used a filter technique for IN determination. At colder temperatures, there was background interference due to large

blank values of IN on the filters. The results at $-20^{o}C$ prove to be consistent from data set to data set and also had the largest number of good determinations.

In general, the measurement of IN in the atmosphere is a difficult task. There is no absolute method that provides absolute IN conentration data, as is the case for CCN. The method itself introduces scatter in the data which cannot be accounted for. The results of the IN data reflect this and are not as precise as the results for the CCN data. Further, no universally applicable relationship has been found to predict actual cloud ice crystal concentration from measured IN concentrations. However, in a broad sense, a coherent picture does emerge.

The primary objective of the study, to determine the effect of pollutant transport to the Arctic on the IN and CCN concentration, is best addressed by comparing the difference in the seasonal concentrations. The winter to summer contrast in pollutant concentration is large, and the seasonal change in the IN concentration should provide the best estimate of the effect of the presence of pollutants on the IN. Table 5.4 is of the same format as Table 5.2 which was used for the seasonal comparison of CCN to the pollutant tracers. The mean winter and summer conentration of the IN are presented in the first two columns, and the winter/summer ratio in the center column. The last two columns contain the seasonal mean pollutant tracer concentration ratios for each site. These are the same values as in Table 5.2.

The single most significant result which arises from this table of all the mean data is the inverse seasonal relationship between the IN

TABLE 5.4

SEASONAL MEAN CONCENTRATION OF POLLUTANT AEROSOL TRACER COMPONENT AND ICE NUCLEI ACTIVE AT -20^{o} AND 1% SUPERSATURATION

Sampling Location	IN Concentration (IN m^{-3})		Winter/Summer Ratio	Aerosol Winter/Summer Ratio	
	Winter	Summer		Noncrustal Vanadium	Nonmarine Sulphate
Barrow	47	144	0.33	>45	>38
Ny Alesund-YMER	59	80	0.74	11	29
Poker Flat	59	750	0.08	2.5	8.3
Pangnirtung	41	98	0.42	7.7	---
Irafoss	56	44	1.3	0.08	<0.40
Narragansett	402	918	0.44	2.0	1.0

concentrations and the pollutant-tracer concentration. The largest seasonal pollutant winter/summer ratio at Barrow coincides with the smallest IN winter/summer concentration ratio. The smaller seasonal pollutant winter/summer ratio at Ny Alesund-YMER coincides with a similarly, slightly larger winter/summer ratio for the IN concentrations. At the low-Arctic sites of Poker Flat, Pangnirtung and Irafoss the same apparent trend continues. Further evidence of the inverse relationship between pollutats and IN are the results of the study at Irafoss. This location had the highest pollutant concentrations during the summer, as mentioned previously and discussed in detail in Appendices C and I. The mean IN concentrations of the summer reflect this by being lower than those of the winter. This evidence suggests that an enhanced removal of IN during winter as a result of colder temperatures activating and causng 'snowout' of the IN may not be responsible for lower concentrations during winter. This cold temperature effect is discussed in detail later (see Fig. 5.10 and the associated discussion).

For the same reasons as discussed in the section on CCN and because of a local dust source of IN during summer, the Pangnirtung data are not considered to be highly significant, but do reflect the same trend. Poker Flat, the continental site, had the largest seasonal change in IN concentration, exceeding a factor of ten from winter to summer. The winter/summer ratio for Poker Flat was not calculated using the one sample during the summer where IN concentration exceeded 10^4 m^{-3}. This is discussed in detail in Appendix G.

The mean overall IN spectra for each experiment site is plotted for the summer and winter seasons in Figures 5.8 and 5.9. It is obvious

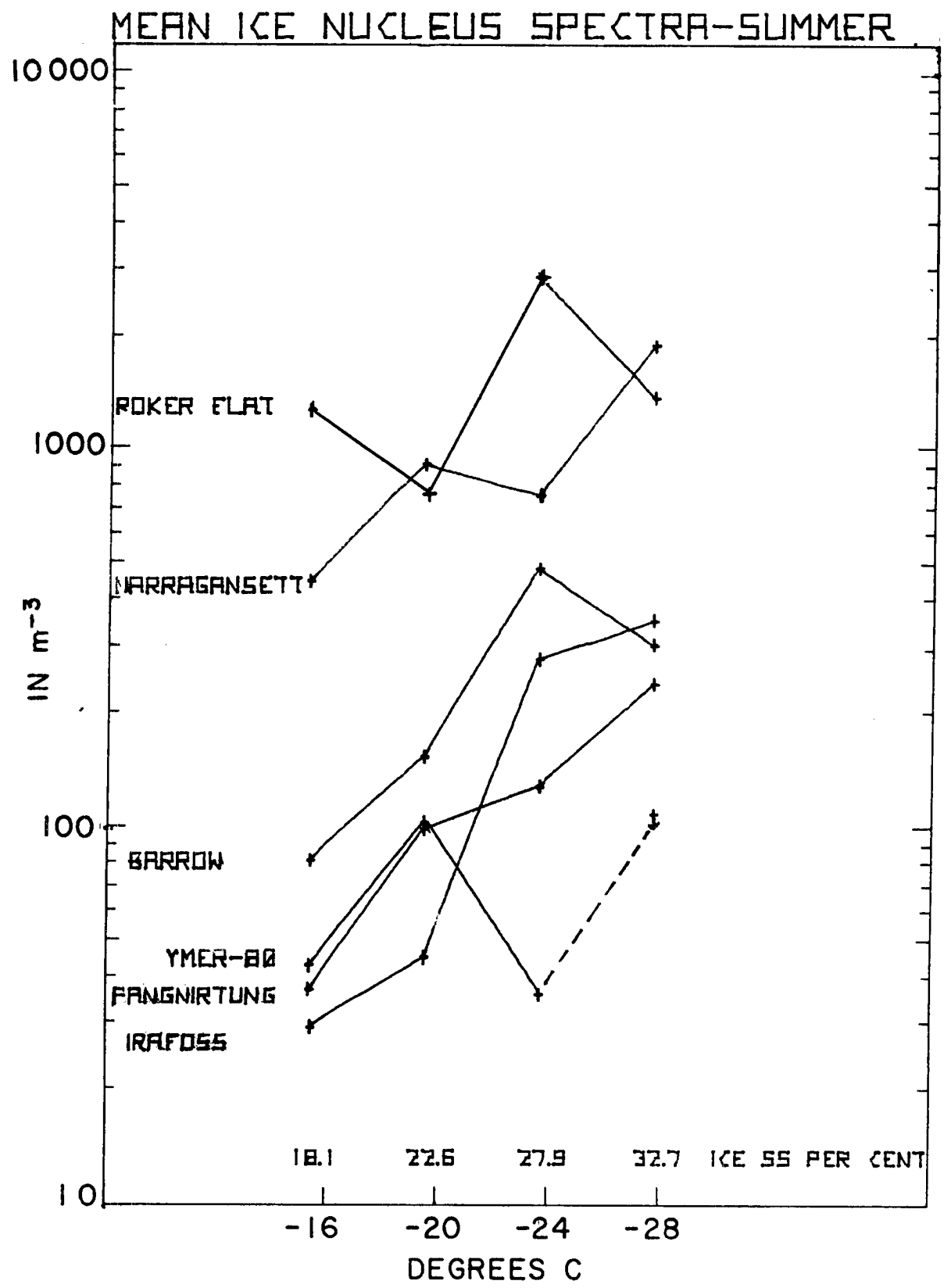


Fig. 5.8 Summary plot of the mean summer IN spectra for the five arctic aerosol experiment sites and the one mid-latitude source region site.

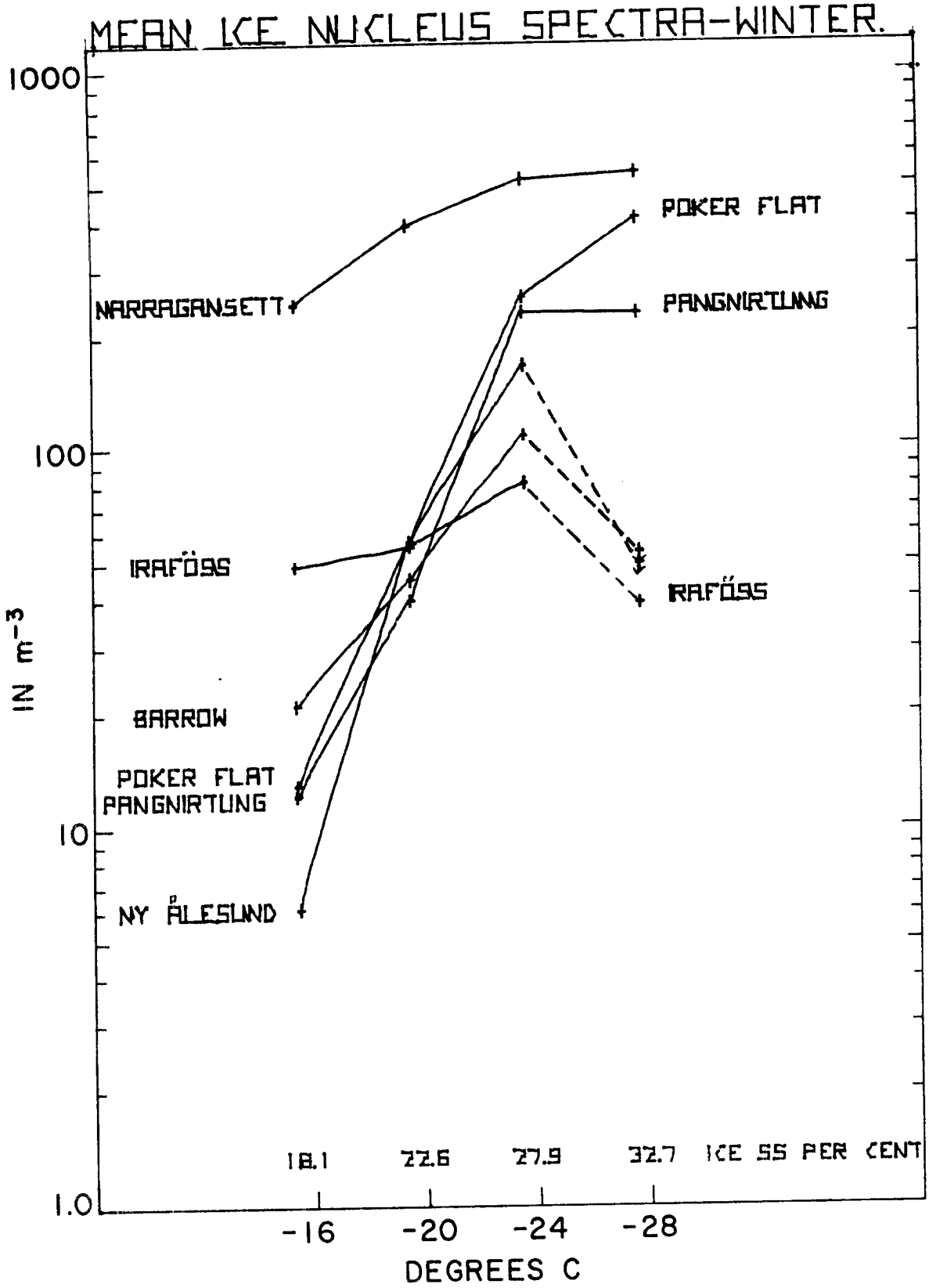


Fig. 5.9 Summary plot of the mean winter IN spectra for the five arctic aerosol experiment sites and the one mid-latitude source region site.

from these plots that there is more scatter in the data than there was in the case of the mean CCN spectra. The individual data points for each sample used to derive the mean spectra are plotted on the mean IN plots in the respective appendices for each site.

The spectra for the winter, pollution-affected arctic sites are quite similar. Irafoss, the only arctic site with more pollutants present during summer, stands out as having a significantly different slope. The IN spectra do not separate themselves in a ranking which is similar to the concentration ranking of the pollutant aerosol tracers, as was found for the CCN. The absolute concentration of IN at any given activation temperature at a particular site is apparently dependent on many variables which vary from site to site Speculation on what factors may be important at the different sites are discussed in the appendices.

The winter/summer contrast in the IN mean spectra is apparent in Figures 5.8 and 5.9. The most dramatic change occurs for the continental arctic site, Poker Flat. However, because the local environment at each site may be important in understanding the IN concentration and their variability, detailed attempts to explain the observations should primarily be restricted to data sets collected at the same location. An overall unified picture of the dependence of the IN on the aerosol tracers at an individual site does not emerge as it did for the CCN data. The one apparent similarity between the data which remains is the inverse winter/summer relationship of the IN with the mean pollutant aerosol tracers.

The observed lowered IN concentrations with elevated pollutant aerosol concentrations are primarily a result of the contrast between the seasons. Table 5.2, which summarizes the time series dependency

between the IN and the aerosol tracers, shows that the inverse relationship with respect to the pollutants from the seasonal data does not necessarily occur on a day-to-day basis. Only a third as many significant correlations occur between IN and the aerosol source tracers as occurred between the CCN and the tracers. Positive significant correlations were observed between the IN concentrations and one or both of the pollutant aerosol tracers for the experiments at Barrow, YMER-80 and Naragansett. As in the case for the CCN, the strong positive correlation of IN with nonmarine sulphate on the YMER expedition is a result of the first three samples collected south of the polar front in an airmass affected by transport from Europe. The data from these three field experiment sites show that within a given airmass containing pollutant aerosol, the IN co-vary with the pollutants and not the natural aerosol. However, for the same location, when pollutant concentrations are lower, and the overall aerosol mass loading is lower, there are more IN present. The IN within a polluted airmass are more closely linked to the man-made aerosol than the natural aerosol in the same airmass but overall, the absolute concentrations of the IN in the polluted airmass are lower.

The results from the Poker Flat winter experiment support these observations in a unique way. In Table 5.2, the only significant IN correlation in the time series data at this site was with noncrustal vanadium. However, this correlation is negative, unlike the other experiment results. As mentioned previously, the Poker Flat site is separated meteorologically from the true arctic airmass by the Brooks Range of Northern Alaska. Intrusions of cold arctic air into Central Alaska along with the pollutants, can occur. Characteristically this

air is more polluted than the air which it displaces in central Alaska. This intrusion also brings air with lower IN concentrations than those which existed in the airmass characteristic of Central Alaska. This results in an inverse relationship between IN and the pollutants at this location, whereas 600 km to the north at Barrow the opposite is true. Thus even though the results of the correlation between IN and noncrustal vanadium at Poker Flat and Barrow appear to be inconsistent, upon closer scrutiny it would have been expected. This is discussed in more detail in Appendix G.

The inverse seasonal relationship between IN and the pollutant aerosol tracers suggests the pollutants may be inhibiting the ability of existing IN by a poisoning effect resulting from chemical or physical interactions between the IN and coexisting pollutants. One possible alternative explanation for this observation may be a selective removal of IN from the atmosphere by the natural snow-forming process which occurs as a result of the nucleation of ice in clouds by the IN present. This process is more efficient at colder temperatures. More numbers of IN will be activated, and removed at colder temperatures. This process may partially explain why winter IN concentrations are lower than summer IN concentrations in the Arctic.

In an effort to address why there is a seasonal difference in IN concentrations, all of the IN concentration data at $-20^{o}C$ are plotted versus the ambient atmospheric temperature at the time of collection of the IN in Figure 5.10. If atmospheric temperature is responsible for a preferential removal of IN by "snowout", lower IN concentrations would be expected at lower temperatures. The data in Figure 5.10 are plotted by letters indicating the experiment site (see Fig. 5.10 legend). The

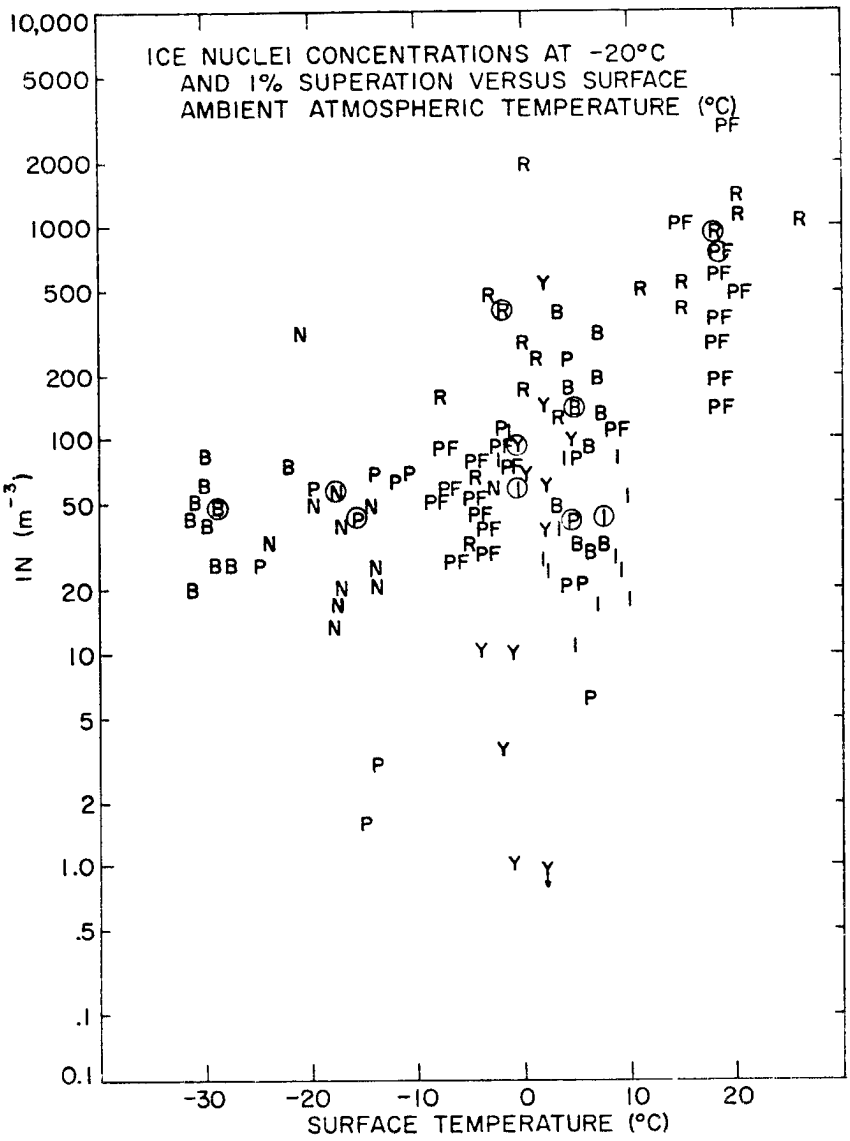


Fig. 5.10 Plot of all ice nuclei concentration data versus surface ambient atmospheric temperature during the collection of the ice nuclei at all field experiment locations. Plotted letters indicate the site where the data was collected. Circled letters indicate the mean values of ice nuclei concentration and temperature for a given site experiment. B = Barrow, AK; PF = Poker Flat, Alaska; P = Pangnirtung, Canada; I = Irafoss, Iceland; N = Ny Alesund, Spitzbergen; Y = YMER-80.

mean temperature and IN concentration for each site is indicated by a circled letter. Lower concentrations of IN do occur at lower ambient temperatures. The greatest change in IN concentrations occurs for temperatures above $0^{o}C$. Below $0^{o}C$ there no longer appears to be a significant decrease in IN concentrations. The data appear to support the hypothesis that selective removal of IN may be partly responsible for the observed seasonal IN concentration differences. The data from Irafoss showed that IN concentrations were lower during the warmer, more polluted, summer experiment. It is apparent that some combination of the selective removal of IN as a result of cold winter temperatures as well as a possible pollutant poisoning effect could explain the observed low winter IN concentrations found at most of the arctic sites. The combination of these two IN removal and inhibition processes working simultaneously may explain why the concentrations of IN measured in the Arctic are amongst the lowest measured anywhere in the world. Excluding the Poker Flat summer experiment, the average IN concentrations at all the arctic sites ranged from a factor of 10 to 50 less in concentration than values reported world wide in the laterature (Purppacher and Klett, 1980). This appears to be unique to the Arctic, and may be related to the highly secondary nature of the aerosol particles and the time and distance over which the particles had been transported from the souce regions in midlatitudes.

Figures 5.11 through 5.15 are scatter diagrams of all of the arctic IN and aerosol tracer data. The same plotting scheme is used here as was used for the CCN data. The dashed line is the best fit linear regression of the logarithms of the data.

Figure 5.11 shows that a negatively sloped regression line best fits the data. The correlation coefficient between the IN and nonmarine sulphate concentrations -0.094, significant at<80% level. There is a large amount of scatter at the lower concentrations of nonmarine sulfate which is not evident at higher concentrations. The pattern does not show any tendency for high IN concentrations at elevated nonmarine sulphate concentration levels as was the case for the CCN.

Figure 5.12, a scatter diagram of all arctic IN versus noncrustal vanadium concentration data shows a similar result. The correlation coefficient for the regression line is -0.247, significant at the 92% level. This evidence is in support of the overall lower IN concentrations found in the arctic with elevated air pollutant concentrations and cold teperatures in winter and vice versa during summer. The combined results of both Figures 5.11 and 5.12 is similar to the results of the mean IN data presented in Table 5.4. The percent of the variance of the IN explained by the pollutant aerosol is small. The most significant result of this data set may be the lack of any evidence of midlatitude pollutant sources of IN in the Arctic.

Figures 5.13 through 5.15 are scatter diagrams of the arctic site IN concentrations data plotted versus aluminum, marine sodium and iodine. None of these diagrams shows a significant relationship between the IN and any of the natural IN tracers used. This was unexpected for the aluminum data since most investigations have shown that natural dust particles, which aluminum was chosen to be an indicator of, are commonly found in ice crystals, and have been shown to have the ability to nucleate ice in the laboratory. This may result from an explanation similar to one given previously for the observed positive correlation

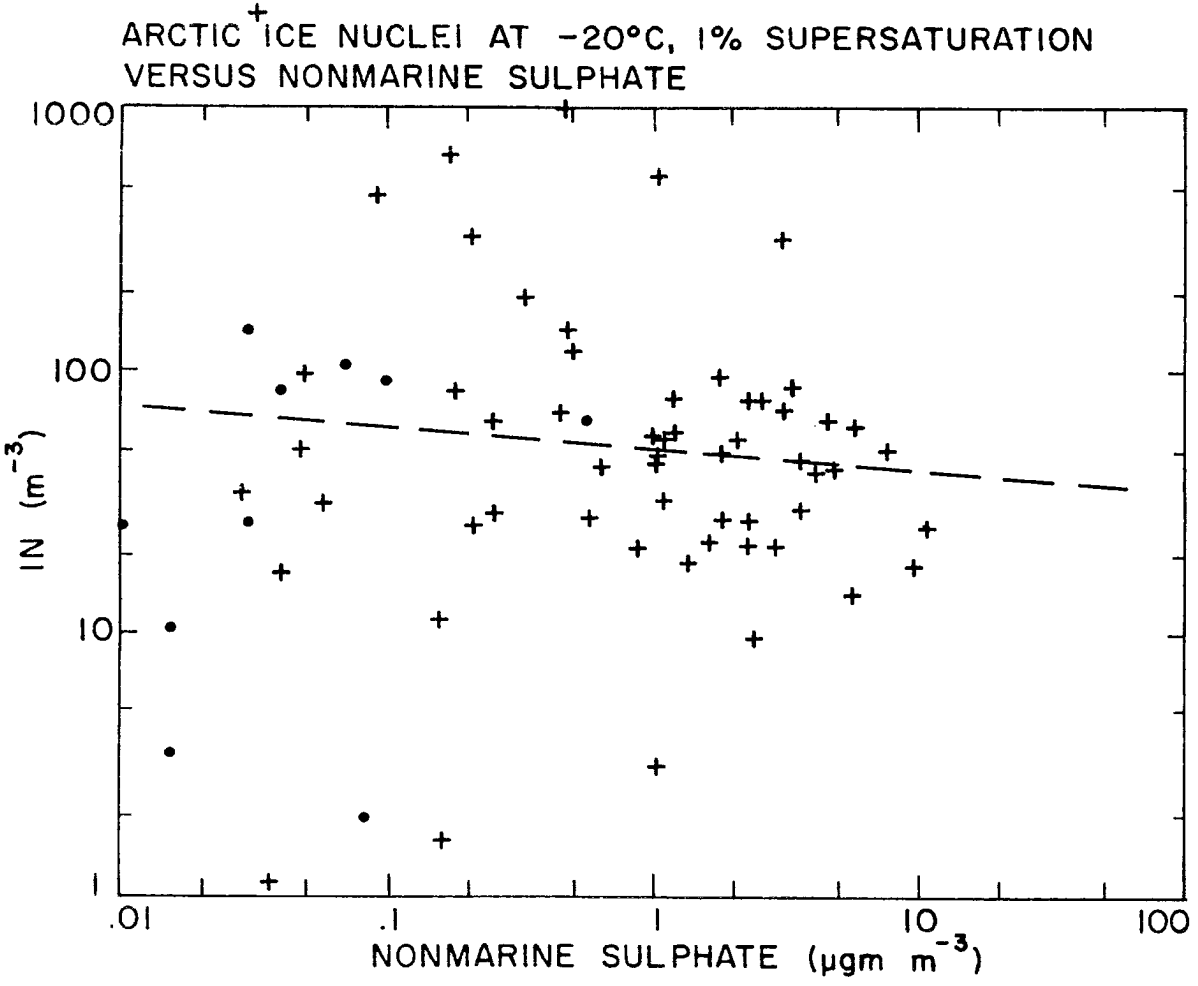


Fig. 5.11 Scatter diagram plot of IN concentrations vs. nonmarine sulphate aerosol concentrations from all five arctic field site experiments----same as Fig. 5.3.

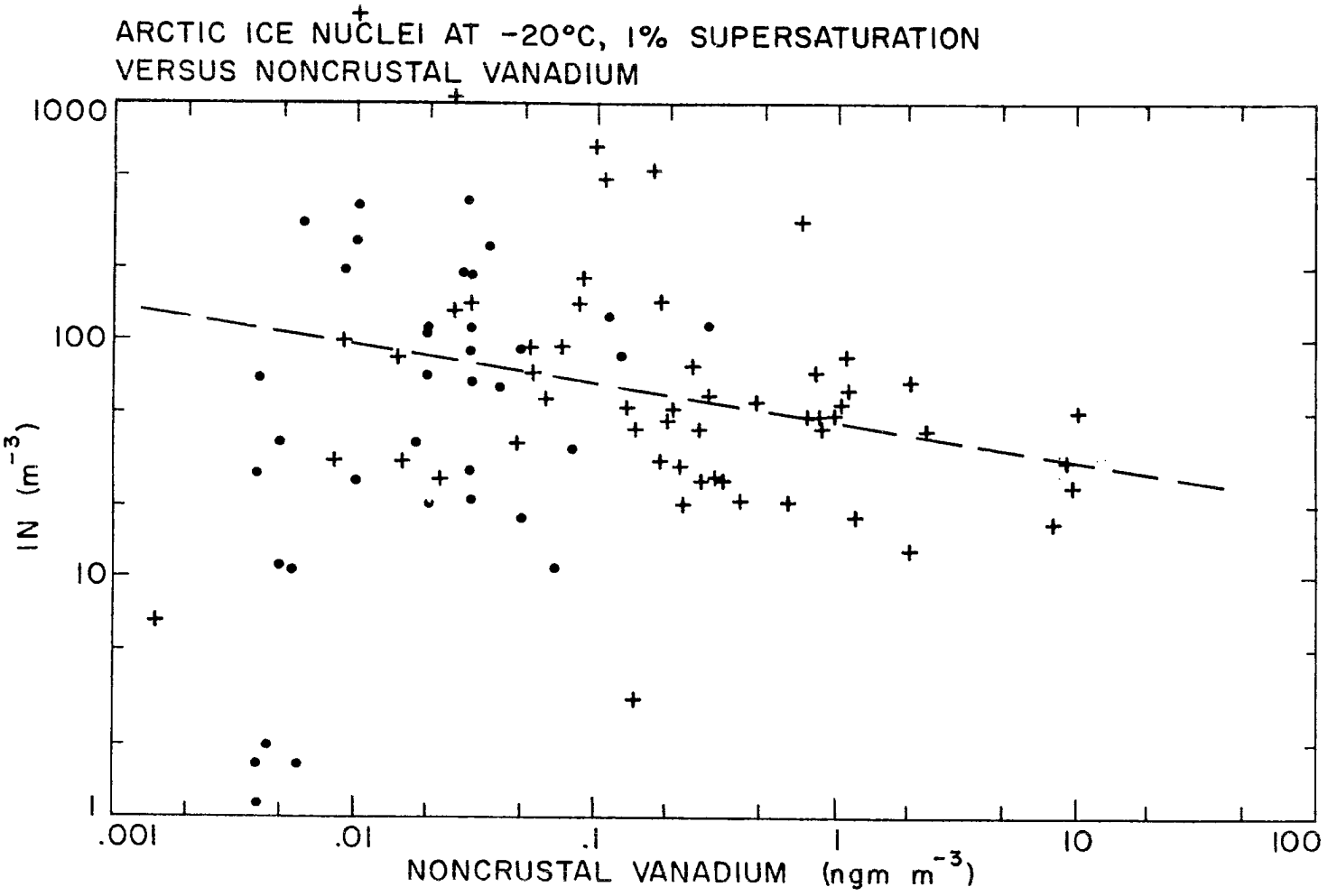


Fig. 5.12 Scatter diagram plot of IN concentrations vs. noncrustal vanadium----same as Fig. 5.3.

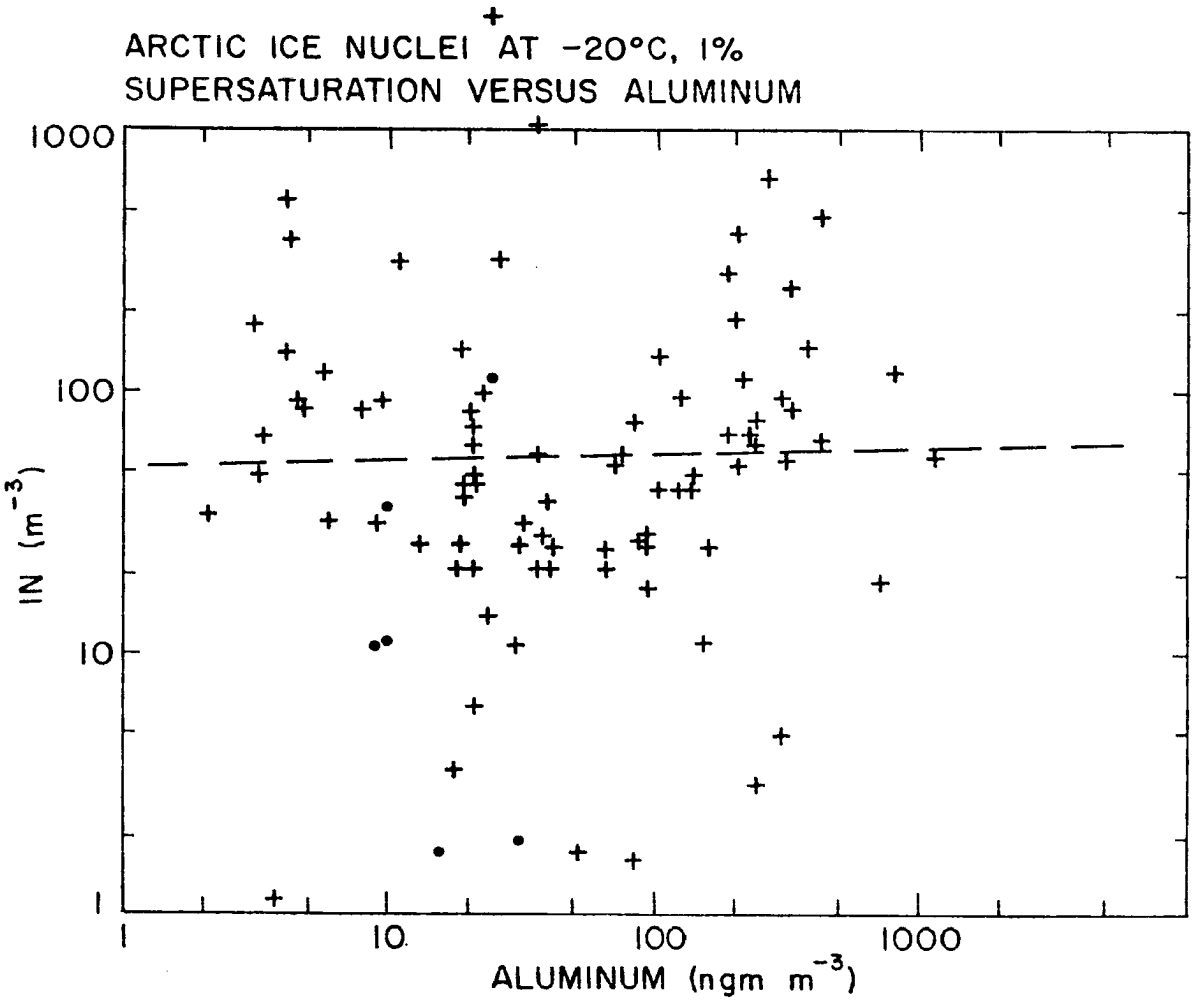


Fig. 5.13 Scatter diagram plot of IN concentrations vs. total aluminum----same as Fig. 5.3.

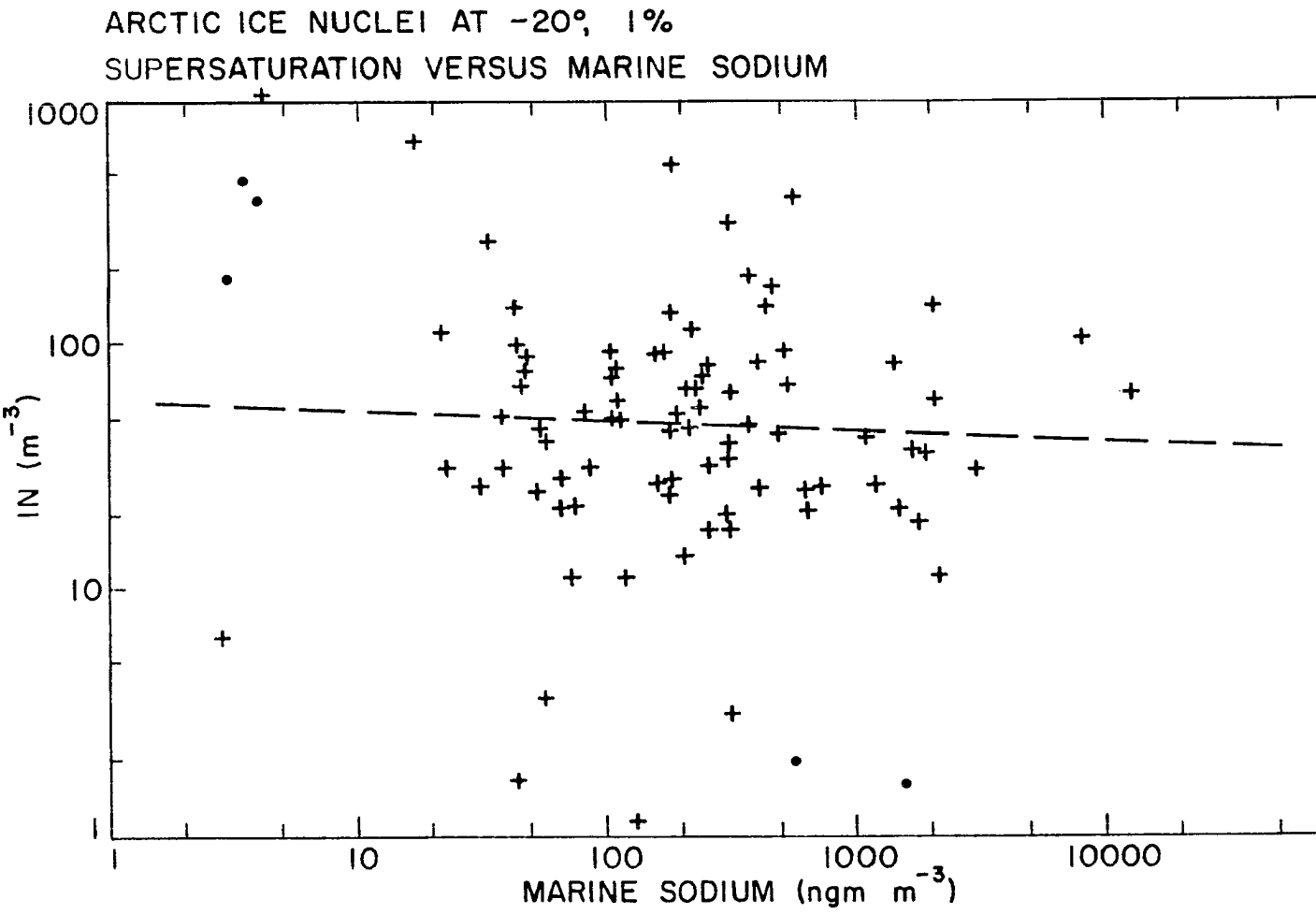


Fig. 5.14 Scatter diagram plot of IN concentrations vs. marine sodium---same as Fig. 5.3.

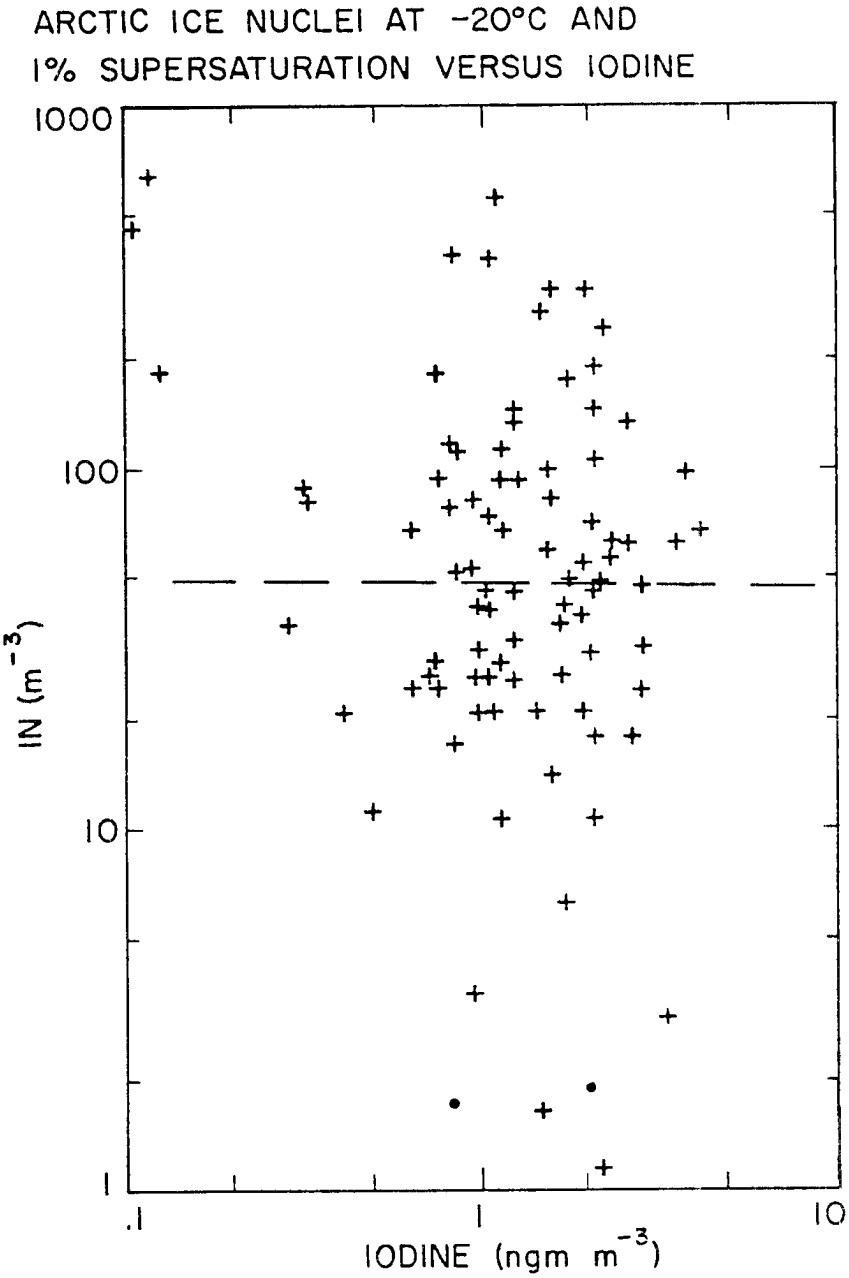


Fig. 5.15 Scatter diagram plot of IN concentrations vs. total iodine --same as Fig. 5.3.

between CCN and aluminum as shown by Figure 5.3. That is, it is possible that aluminum-containing particles such as fly ash associated with the other pollutant aerosol tracers are also present. The relationship between the pollutant aluminum and IN may be opposite that of the aluminum from natural sources. It is impossible to separate the two forms of aluminum in the data set. The net result of such a combination might mask the two coincident positive and negative relationships between the IN and aluminum concentrations, and create the observed result in Figure 5.13. The same result is obtained if only the IN and aluminum data are plotted and analyzed for periods when the pollutant aerosol tracers are present.

The primary objectives of the arctic cloud-active aerosol study have been achieved. The results of the effects of long-range transport of air pollutants on the arctic cloud-active aerosol have been summarized. The CCN and IN concentrations in the Arctic, especially in the true arctic airmass, can be linked to either the day-to-day or seasonal variability of the pollutant aerosol concentrations. A general relationship of this nature did not occur between the cloud-active aerosol and the natural aerosol tracers used in this study. The significance of this result, for a region as large and as climatically importat as the Arctic, of man's influence on the cloud-active aerosol concentrations, will be discussed in the next chapter. In addition, conclusions which can be drawn on the source regions and pathways of the pollutant aerosol based upon these results will be presented.

VI. CONCLUSIONS AND DISCUSSION

This study brings together for the first time the chemical and nucleational characteristics of the arctic aerosol and combines these observations to produce a more complete understanding of their interrelationships. This was done primarily to address the effects of the well-known large anthropogenic component of the total arctic aerosol chemical composition, on the nucelational properties of the arctic aerosol. The interaction of the Arctic Haze aerosol with liquid and vapor phase water may prove to be the most significant environmental effect of this aerosol in the Arctic.

6.1 Cloud Condensation Nuclei and Pollutant Aerosol in the Arctic

The results of simultaneous measurements of pollutant aerosol tracers and CCN in the Arctic show conclusively that pollutants emitted thousands of kilometers away, reach the Arctic, and contribute to the number of CCN in the arctic atmosphere. This finding is true at opposite sides of the Arctic, and is not related to emissions of pollutants within the Arctic. Similar relationships between natural aerosol source tracers and CCN were not found. This in itself is significant. It is possible, based upon the chemical composition of a bulk aerosol sample, to determine what source fraction of that aerosol is primarily responsible for an observed physical property of the aerosol, in this case the ability of the aerosol to form cloud droplets under the proper conditions. In addition, this was accomplished in locations thousands of kilometers from any major pollution sources.

Aerosol elemental and trace constituent analysis can be a powerful tool in other aerosol experiments where knowledge of the source and chemical composition of the aerosol is known.

The results of this study are not unusual. This investigation was concerned with the nucleational properties of Arctic Haze aerosol. By definition, Arctic Haze aerosol contains particles which form haze droplets in the atmosphere. These droplets have been observed by others, and have been attributed to the presence of sulphuric acid in the atmosphere, hypothesized to be a major component of the Arctic Haze aerosol. The same hygroscopic or acidic particles which form haze droplets would form cloud droplets if suitable supersaturations occur. These particles can be considered as CCN.

The transport of particulate pollutants over the distances required to observe them in the Arctic permits only those species which have relatively long residence times in the atmosphere to survive. Basically two prerequisites must be met. The particles must either be of a small size, or they must form along the way as a result of gas-to-particle conversion. The two pollutant tracers used, noncrustal vanadium and nonmarine sulphate fall within these two categories, respectively. The nonmarine sulphate may have originated from the oxidation and hydrolysis of gases such as H_2S or SO_2 as well as having been emitted as a primary aerosol. The sulphate products may be ammonium sulphate or sulphuric acid. Both of these are hygroscopic and would condense water readily. Thus the fact that these pollutants act as good CCN is not surprising, and the strong correlation observed should be expected. The mass of nonmarine sulphate is sufficient to produce the observed numbers of CCN.

Probably the most surprising result of the arctic CCN data was the apparent lack of any strong marine sources of CCN. A closer look at the relative mass concentration of marine sodium versus nonmarine sulphate shows that at all the arctic sites during the winter, the nonmarine sulphate concentrations exceeded the marine sodium concentrations. At Irafoss, where the marine sodium exceeded the nonmarine sulphate during winter, the CCN concentrations were correlated significantly to both of the marine aerosol tracers, marine sodium and iodine (see Table 5.2). It is difficult to determine if this is also true during the summer because of the low aerosol concentrations and the resulting lack of data. This implies that in regions where nonmarine sulphate is present in concentrations greater than the marine aerosol, and the nonmarine sulphate is anthropogenic in origin, man's activities may be significantly altering the CCN concentrations. The extent of this effect may be quite large in the industrialized Northern Hemisphere. In the Arctic the nonmarine sulphate is the major source of CCN.

6.2 Ice Nuclei and Pollutant Aerosol in the Arctic

The results of the IN measurements in the Arctic, are not as conclusive as those of the CCN measurements. The evidence from this data set cannot separate the proposed combined effects of pollutant poisoning of the IN and the selective removal of IN by cold, winter temperatures on the observed low, winter IN concentrations. The cold-winter versus warm-summer temperature effect discussed in Chapter V no doubt plays a role. Evidence presented in Chapter V also suggests that poisoning of the IN by pollutants may also be important.

Ice nuclei initiate the formation of ice in the atmosphere by essentially three different mechanisms. 1) By diffusional growth of ice

directly from the vapor phase (deposition). 2) By freezing of water which has condensed on the IN (condensation-freezing). 3) By freezing of a prexisting supercooled droplet when the IN comes in contact with it. Pollutants could effect all of these mechanisms with the end result being fewer numbers of IN with the ability to nucleate the ice phase.

For the high-Arctic sites, Barrow and Ny Alesund, the concurrent measurement of the CCN and the CN concentrations showed that on the average, during winter when the pollutant aerosol was present, 70%-80% of the total aerosol number concentration, as indicated by the CN concentrations, were active as CCN at 0.2% supersaturation. At the peak concentrations of nonmarine sulphate, the concentrations of CN and CCN were identical. This suggests that all of the aerosol particles were hygroscopic. Yet inspection of the photomicrographs in appendices F and J reveal insoluble particles, and the presence of elements such as aluminum, vanadium and other metals suggest insoluble aerosol is present. For an IN to function it must be insoluble long enough to initiate the ice phase. If all the aerosol particles were coated with a hygroscopic layer of material, sulphuric acid for instance, this may seriously interfere with the ability of the particle to act as an IN. The depositional mode of nucleation could be inhibited by a layer of water on its surface, possibly a concentrated salt solution. The salt solution, if acidic may alter the surface structure of the particle as a result of chemical etching. This could, in turn, inhibit the nucleation of ice by the condensation-freezing mechanism by freezing point depression. Finally, if all the particles are coated with hygroscopic material, and act as CCN forming liquid solution droplets, the contact-freezing mechanism would be eliminated since no dry IN would exist. All

or part of these possible interactions of pollutants with IN could explain part of the observed lowering of IN concentrations when the pollutants are present. More detailed studies would be necessary to determine whether the poisoning hypothesis is valid and what fraction of the total lowering of the IN conentrations it can explain.

As was the case for CCN, no indication of a natural source of IN could be found for the Arctic in general. This is based upon the scatter diagrams, Figures 5.13-5.15 which present all of the IN data. However, the IN did show some relationship to natural sources at individual sites such as marine sources at Barrow during summer (see Table 5.2 and Appendix F) and to local dust at Pangnirtung during summer (see Table 5.2 and Appendix H). Poker Flat had the highest IN concentrations measured out of all the sites including Narrangasett. These high concentrations occurred during the summer experiment, yet they did not correlate with any of the aerosol tracers measured. These results provide evidence that dust, marine sources (possibly biogenic) and an unknown source (also possibly biogenic) of IN exist at these locations. It was not possible to determine what these sources were from this data set. The use of additional tracers may provide more clues in future studies. This study points toward the need to further understand what are the natural sources of IN in the atmosphere in general, and the Arctic in particular, and what ice nucleation mechanisms are important for each source, in order to thoroughly understand how man-made pollutants may effect the ability of the IN to form ice in the atmosphere.

6.3 Effects of Pollutant Aerosol on Arctic Clouds

The basic microstructure of clouds depends upon the aerosol particles in the atmosphere which nucleate liquid water and ice. Knowledge of the atmospheric concentration of these nucleants allows an estimate to be made of the microstructure of a cloud which forms in the airmass containing the nuclei. Cloud condensation nuclei are responsible for the formation of liquid cloud droplets. Ice nuclei initiate the freezing of these droplets or possibly form the ice phase directly from water vapor. In stable-stratiform clouds, particularly at cold temperatures ($<-20^{o}C$) it is possible to predict cloud droplet number concentrations and ice particle concentrations if the CCN and IN concentrations are known. This is more accurately done for cloud droplet formation, and prediction of their concentration. Ice crystal concentration prediction is not as straightforward, and may result from the difficulties in measuring IN concentrations in the atmosphere. However, some success has been shown to be possible.

Clouds in the Arctic are primarily stratiform. The application of measurements of CCN and IN concentrations in the arctic atmosphere to estimate actual in-cloud droplet and ice crystal concentrations is certainly possible. The results of this study have shown that the CCN concentrations respond positively to increasing pollutant aerosol concentrations as pollutants are transported into the Arctic. Both daily and seasonal increases of CCN concentrations by factors of 5 to 8 were observed as pollutant laden airmasses either displaced a more natural, background airmass during a site experiment or were present during the winter in general, in contrast to the summer. Even though the IN measurements were not as precise as those for CCN, concentrations

of IN were consistently lower by factors of 2 to 3. This was primarily a result of the winter/summer comparisons at the arctic sites.

The effects observed on the nucleant concentrations in the arctic atmosphere should be reflected in the microstructure of arctic stratiform clouds. Clouds which form within airmasses which are under the influence, or contain pollutants from, mid-latitude source regions at any time of the year, would be expected to have higher cloud droplet concentrations and lower ice crystal concentrations than clouds in the same region which form in a more representative background-arctic airmass. For this prediction to be valid for cloud ice-crystal concentrations, heterogeneous ice nucleation mechanisms would have to dominate the formation of ice crystals be homogeneous nucleation. The exact difference in the number concentration of droplets and ice crystals between the polluted and pristine clouds would depend upon the relative concentration of pollutants. The greatest difference in cloud microstructure would result from the changes in cloud droplet concentrations. This results from the larger chages which occurred in the CCN versus the IN concentrations, and because IN concentrations are typically 10^3 to 10^5 less than the concentrations of CCN. Thus the dominating effect of pollutants on arctic cloud microstructure would be in the increased number of cloud droplets formed.

This study was conducted entirely at ground level. It is not known how respresentative these measurements may be of IN, CCN and pollutant concentrations at the levels in the atmosphere at which clouds form. The representativeness of these ground studies are a critical question which must be answered in the future.

6.4 Pollutant Pathways to the Arctic

The primary objective of determining whether pollutants transported to the Arctic effected the concentration of cloud-active aerosol additionally provided information on the transport pathways of the pollutants to the Arctic. This was the primary reason for the selection and location of the low-Arctic sites of Poker Flat, Pangnirtung and Irafoss as described in detail in Chapter IV.

Poker Flat, lying 600 km south of Barrow, was chosen to determine whether any measurable pollutants at Barrow might arrive as a result of transport over the North Pacific Ocean, form East Asia and Japan. Transport from the deserts of Asia to Alaska had been reported previously (Rahn et al, 1977b). Pangnirtung was chosen to lie along the potential transport pathway from the east coast of North America along the west of Greenland, as suggested by Radke et al. (1976), to the north and west to Barrow. Irafoss was chosen to lie along the proposed pathway from the east coast of North America, across the North Atlantic, and to the north on the east side of Greenland. In addition to the low-Arctic sites, Barrow and Ny Alesund-YMER were used to provide information on the North Pacific transport pathway (Barrow) and on the transport pathway from the east coast of North America, Europe and Eurasia (Ny Alesund-YMER). These pathways are depicted in Figure 4.1.

The results from the Poker Flat studies (detailed in Appendix G) never indicated that transport from the south at that location was ever associated with elevated pollutant aerosol tracer concentrations resulting from long-range transport. On the contrary, the highest concentrations of noncrustal vanadium at Poker Flat were associated with flow at low levels from the northeast, within a cold-polar-arctic

airmass. Flow from the south had concentrations approximately a factor of 8 lower. This is in agreement with investigations by Rahn (personal communication) from a site near Fairbanks, Alaska which were representative of a long period of study.

The elimination of the North Pacific transport pathway of pollutants is understandable in light of the storminess of the region and the efficient removal of aerosol particles. The transport of Asian desert dust to Alaska was accomplished by the combination of a massive short-term dust source, rapid mixing in the vertical, and transport at high altitudes, apparently above the precipitating regions of the atmosphere. This event is also the exception rather than the rule, and even if pollutant transport were associated with it, it could not explain the observed high pollutant concentrations in the Arctic over the course of the winter.

Sampling at Pangnirtung revealed no significant transport of pollutants from North America along the west side of Greenland. The pollutant concentrations were elevated during the winter experiment relative to the summer experiment. The summer experiment pollutant concentrations were at or below their detection limit, whereas the winter experiment values were slightly above the detection limit. It is possible that a longer experiment at this location might provide further evidence of possible pollutant-transport events from North America. The concentrations of pollutants observed were too low to account for the winter concentrations further north. As detailed in Appendix H, the pollutant concentration gradient for noncrustal vanadium at the time of

the winter experiment was positive to the north. This does not support the hypothesis of pollutant transport from the south at this location.

Pangnirtung was downwind of the North American source region during the winter experiment. Transport of airmasses were associated with low pressure centers off the east coast of North America as they traveled to the east and north. The North Atlantic Ocean at this time is extremely stormy and, much like the airmasses of the North Pacific Ocean, efficiently remove aerosol particles as a result of precipitation and cloudiness. The meteorology of this transport pathway is not conducive to long-range pollutant transport from North America. Further evidence of this was obtained at Irafoss.

Irafoss, Iceland is located approximately half-way between North America and the High Arctic. During the winter experiment, transport from the direction of North America was very rapid. It is estimated from the wind speeds at the time that transport times from North America to Irafoss may have been on the order of 2-3 days. However, no significant concentrations of the pollutant tracers were measured. the concentrations of noncrustal vanadium were 10 to 100 times lower at Irafoss during winter than at the high-arctic sites of Barrow and Ny Alesund. For the same reasons as those given for Poker Flat and Pangnirtung, the aerosol transported off the coast of North America cannot survive the efficient removal processes associated with the stormy North Atlantic.

Irafoss was unique in having a summer maximum in pollutant tracer concentrations which was very important in understanding the relationship of IN with the pollutants in the Arctic (see section 6.2). The summer observation was in the form of an event of transport from

Europe and not representative of an average condition. Although these events occur often during the summer season, they play no role in determining the aerosol chemical composition of the high-Arctic atmosphere. They are an example of long-range pollutant transport affecting a region with no pollutant emissions of its own.

Although the furthest north of all the arctic sites (79°N), Ny Alesund is also situated in a location that places it along several potential transport pathways from Europe and Eurasia as well as North America (see Fig. 4.1). The North American pathway has been eliminated by the observations at Irafoss, approximately 2000 km further upwind. The Europe-Eurasian pathways appear to play highly significant roles in the determination of the aerosol chemical composition of the winter arctic atmosphere.

The concentrations of pollutants measured at Ny Alesund were the highest of all the arctic sites. This is not unusual, since it is located relatively close to the highly industrialized regions of Europe and the western U.S.S.R. The advection of the pollutants and the meteorology of transport are discussed in detail in Appendix J. The transport along this pathway from central Europe to Scandanavia has been well known for years as a result of acid precipitation studies in the Scandanavian countries. The measurements made at Ny Alesund confirm that the transport continues to the north after leaving the north coast of Europe-Eurasia. The pathway is very efficient because the sources are so near the arctic. Central Europe is located at approximately 50-55° N. Removal processes do not have a great deal of time to operate. The meteorology is conducive to this transport on a regular basis during winter. This is a result of the semi-permanent position of the

Icelandic Low to the west and the high pressure of the cold continent to the east. The mean southerly flow which results, lies over the major pollutant emission regions of this highly industrialized area. This transport pathway is not dominated by a stormy, low pressure region. The additional influence of the cold, high pressure to the east over the continent permits some portion of the transport to be associated with dry, nonprecipitating conditions. This is most likely the single most important factor responsible for the observation of pollutant transport from Eurasia well into the Arctic. It is highly probable that transport along this pathway, and other pathways from the interior of the U.S.S.R. which were not involved in the pollutants observed during the experiments described here, play a major role in accounting for Arctic Haze aerosol.

6.5 Pollutant Source Regions for Arctic Aerosol

It becomes readily apparent from the previous discussions that the industrialized regions of Eastern Asia (Japan, China, and eastern parts of the U.S.S.R.) and North America are not responsible for emissions of pollutants which can be transported to the high-Arctic. No evidence of transport from these areas was found. The chemical and meteorological evidence points toward Europe-Eurasia as most likely, if not certainly, responsible for much of the pollutants observed in the arctic atmosphere. The major unknown is what fraction of the observed pollutants are emitted directly within the Arctic as a result of man's activities in the U.S.S.R., or transported further north from more southerly regions in the U.S.S.R. Emissions from these southern areas cannot be ruled out as a result of the data presented in this study. This is the single largest unknown at this time concerning the source

region most responsible for the pollutants in the Arctic, and the subsequent effects on the cloud-active aerosol concentrations.

A large amount of smelting, mining of coal and iron production is done in north central U.S.S.R. and further south in the region around the Ural mountains. Much of this activity lies north of 55°N. Situtated well within the cold-dry-continental high pressure system which dominates this region in the winter, the emissions can occur directly into an arctic airmass virtually devoid of any significant wet removal processes. If this airmass is advected to the north, it may contribute significantly to the total pollutant aerosol loading of the winter arctic atmosphere. This is only speculation at this time, but refinement of the tracer techniques using the aerosol trace chemical composition may soon allow source type discrimination within a region to be done on a routine basis, thousands of kilometers downwind (Rahn, personal communication). Since direct measurements of the aerosol are not possible in the Eastern Arctic source regions, this technique will prove invaluable in ascertaining the relative roles of Europe, Eurasia and the U.S.S.R. as source regions for the winter arctic aerosol and its effects on the CCN and IN concentrations of the arctic.

6.6 Global Implications of This Study

The results of this study have more application than just to arctic cloud physics and arctic atmospheric chemistry. The findings certainly have global implications. These results show that the effects of man-made pollutant emissions can be readily measured at distances approaching 5000 km away from their sources. Most importantly the effects of these emissions are wide-spread in the Arctic covering an area centered on the north pole 40° to 50° latitude in diameter. The

measured effects now include altering the climatically important cloud-active aerosol concentrations. These nucleants are responsible for cloud microstructure, how the cloud interacts with the solar and terrestrial radiation fields, as well as the precipitation efficiency of the cloud. The climate of the Arctic is an important link in the overall global climate. Significant changes in the climate of the Arctic have been postulated by many scientists as potentially having far reaching global effects. The magnitude of the possible effects can only be speculated upon. The nucelational properties of pollutant emissions by man may prove to be one of the greatest perturbations on the atmosphere because of the changes in cloud microstructure which may result and the implications of these changes on the regional and possibly the global climate. It is necessary to continue to try to understand how man influences his environment with further efforts in this area of atmospheric science.

VII. SUGGESTIONS FOR FUTURE RESEARCH

The experiments, both in the field and in the laboratory were highly successful. The chemical analysis and collection of aerosol samples proved to be routine. Analysis of the CCN by the instrument used was tedious because it was a primary method, and involved the individual counting of the cloud droplets activated. However, the results were very precise and of excellent quality. Measurements of the IN concentrations were the least precise. The method, although used extensively, is still not an absolute technique. This is true of all the current field measurement techniques used to determine IN concentrations. Until an absolute standard method is found, most IN measurements will have to be made and interpreted in a relative sense using one method. In this sense, the IN measurements made during this study are considered to be significant. The overall results of the study were dependent on the quality of the data. Any future measurements of this type should incorporate an IN measurement technique that can be used in real time, much like the CCN measurements. This would undoubtedly decrease the amount of scatter in the IN data by helping to eliminate some of the 'noise' in the measurements.

None of the measurements made were redundant or unnecessary. Any future field experiments of this type may be significantly improved by increasing the aerosol analyses performed This would increase the number of tracers that could be used to narrow down the different aerosol source types which may account for the observed variability in the

cloud-active aerosol concentrations. In particular, the soluble fraction of the aerosol might be analyzed separately from the insoluble fraction for the different major ions. This may help in the study of natural CCN sources. The insoluble fraction may have a more significant relationship with the IN concentration than the total bulk aerosol analyses used in this study. Finally with regard to aerosol chemistry, it would be advantageous to extend the trace elemental composition analysis to include more elements known to be emitted from different source types. Using the method used in this study, neutron activation analysis, this could be accomplished by an analysis scheme which would include the radionuclides with longer half-lives. This would entail another irradiation of the sample, resulting in a doubling of the useful information.

The most pressing need for future research is to confirm or disprove the hypothesis that the surface measurements of the aerosol and the concentrations of the nuclei are applicable at the levels in the atmosphere where clouds form. A future experiment involving simultaneous ground-based and airborne measurements of the CCN and IN concentrations and the chemical composition of the aerosol is needed. Without this information, extrapolation to the free atmosphere cannot be considered warranted.

This is, of course, most necessary for the cloud-active aerosol. It is imperative that actual in-cloud measurements of cloud droplet and ice crystal size distributions and concentrations be performed in conjunction with aerosol chemical composition measurements similar to those done in this study. Many investigations of cloud water chemistry have shown that the cloud water, in the form of liquid droplets,

efficiently scavenges the interstitial aerosol and soluable-reactive gases. It has been claimed that the cloud water would provide an excellent means of collecting a representative sample of the aerosol which was present in the airmass within which the cloud formed. In addition, the cloud itself could be used as the 'chamber' within which the CCN and IN that are naturally important are activated, allowing them to be counted as droplets and ice crystals. This eliminates the uncertainty of what supersaturation, with respect to ice or water, best approximates the real atmospheric cloud condition, and what mechanism of ice nucleation is important. An airborne experiment of this type would be simpler to execute and analyze. Measurements of IN and CCN by instrumental methods would not be necessary. The natural cloud would perform all of the necessary work by concentrating the aerosol in the liquid water drops and activating the CCN and IN. Many variables could be eliminated and the cloud would serve to smooth out and integrate much of the variability that would otherwise pose a problem.

New aircraft measurement systems for real time cloud droplet and ice crystal concentrations exist. Many methods for the collection of cloud water also have been tested and used in other experiments. Combining these in an airborne arctic cloud-active aerosol and cloud microphysics experiment is feasible at this time and has an excellent chance for success. Not only would an experiment of this nature determine whether the results of this experiment are applicable to clouds, but it would serve to answer the question whether ground measurements of the Arctic atmosphere are representative of the arctic atmosphere aloft.

The arctic atmosphere has been studied relatively little. There is a great deal of opportunity for further research in the area of man's influence on the atmosphere as a result of pollutant emission. The winter arctic atmosphere provides a unique opportunity to pursue this research.

REFERENCES

Auer, A. H. (Jr.), D. L. Veal and J. D. Marwitz, 1969: Observations of ice crystals and ice nuclei concentrations in stable cap clouds. J. Atmos. Sci., 26, p. 1342-1343.

Barrie, L. A., R. M. Hoffman and S. M. Duggupaty, 1981: The influence of mid-latitudinal pollution sources on the haze in the Canadian Arctic. Atmos. Env., 15, p. 1407-1419.

Bigg, E. K. and J. Giutronich, 1967: Ice nucleation properties of meteoric material. J. Atmos. Sci., 24, p. 46-49.

Bigg, E. K. and C. M. Stevenson, 1970: Comparison of concentrations of ice nuclei in different parts of the world. J. Rech. Atmos., 4, p. 41-58.

Bigg, E. K., 1980: Comparison of aerosol at four baseline atmospheric monitoring stations. J. Appl. Meteor., 19, p. 521-533.

Bodhaine, B. A., J. M. Harris and G. A. Herbert, 1981: Aerosol light scattering and condensation nuclei measurements at Barrow, Alaska. Atmos. Environ., 15, p. 1375-1389.

Borys, R. D., 1981: Modified Mee Industries static thermal gradient diffusion cloud chamber. J. Rech. Atmos., 15, 267-269.

Braham, R. R. and P. Spyers-Duran, 1974: Ice nucleus measurements in an urban atmosphere. J. Appl. Meteor., 13, p. 940-945.

Brier, G. W. and D. B. Kline, 1959: Ocean water as a source of ice nuclei. Science, 130, p. 717.

Carlson, T. N., 1981: Speculations on the movement of polluted air to the Arctic. Atmos. Env., 15, p. 1473-1477.

Daisey, J. M., R. J. McCaffery and R. A. Gallagher, 1981: Polycyclic aromatic hydrocarbons and total extractable particulate organic matter in the Arctic aerosol. Atmos. Env., 15, p. 1353-1363.

Darby, D. A., L. H. Burckle and D. A. Clark, 1974: Airborne dust on the Arctic pack ice, its composition and fallout rate. Earth and Planetary Sci. Let., 24, p. 166-172.

Dinger, J. E., H. B. Howell and T. A. Wojciechowski, 1970: On the source and composition of cloud nuclei in a subsident air mass over the North Atlantic. J. Atmos. Sci., 27, p. 791-797.

Duce, R. A., J. W. Winchester and T. W. Van Nahl, 1966: Iodine, bromine and chlorine in winter aerosols and snow from Barrow, Alaska. Tellus, 18, p. 238-248.

Duce, R. A., G. L. Hoffman and B. J. Ray, 1975: Atmospheric trace metals at remote northern and southern hemisphere sites: pollution or natural? Science, 187, p. 59-61.

Duce, R. A. and G. L. Hoffman, 1976: Atmospheric Vanadium Transport to the Ocean. Atmos. Env., 10, p. 989-996.

Eagan, R. C., P. V. Hobbs and L. F. Radke, 1974: Measurements of cloud condensation nuclei and cloud droplet size distribution in the vicinity of forest fires. J. Appl. Meteor., 13, p. 553-557.

Egorov, V. V., T. N. Zhigalovskaya and S. G. Malakhov, 1970: Microelement content of surface air above the continent and the ocean. J. Geophys. Res., 75, p.3650-3656.

Fenn, R. W. and H. K. Weickmann, 1959: Some results of aerosol measurements. Geofis. Pura. e Appl., 42, p. 53-61.

Fenn, R. W., 1960: Measurements of the concentration and size distribution of particles in the Arctic air of Greenland, J. Geophys. Res., 65, p. 3371-3376.

Fenn, R. W., H. E. Gerber and D. Wasshausen, 1963: Measurements of the sulphur and ammonium component of the Arctic aerosol of the Greenland icecap. J. Atmos. Sci., 20, p. 466-468.

Fitzgerald, J. W. and D. A. Spyers-Duran, 1973: Changes in cloud nucleus concentration and cloud droplet size distribution associated with pollution from St. Louis. J. Appl. Meteor., 12, p. 511-516.

Fletcher, N. H., 1962: Physics of Rain Clouds. Cambridge Univ. Press, London, Eng.

Fletcher, J. C., 1965: The heat budget of the Arctic Basin and its relation to climate. Report R-444-PR, The RAND Corp., 1700 Main St., Santa Monica, CA 90406, 179 pp.

Flyger, H., K. Hansen, J. Megaw and L. C. Cox, 1973: The background level of the summer tropospheric aerosol over Greenland and the North Atlantic ocean. J. Appl. Meteor., 12, p. 161-174.

Flyger, H., N. Z. Heidam, K. Hansen, W. J. Megaw, E. G. Walther and A. W. Hogan, 1976: The background level of the summer tropospheric aerosol, sulphur dioxide and ozone over Greenland and the North Atlantic ocean. J. Aerosol Sci., 7, p. 103-140.

Georgii, H. W. and E. Kleinjung, 1967: Relations between the chemical composition of atmospheric aerosol particles and the concentration of ice nuclei. J. Rech. Atmos., 3, p. 145-156.

Gergach, A. L., G. M. Zobrodsky and V. G. Morachevsky, 1960: The results of a complex investigation of the type St-Sc clouds and fogs in the Arctic. IZV. Geophys., Ser., p. 100-114.

Griffin, J.J. and E.D. Goldberg, 1979: Morphologies and origin of elemental carbon in the environment. Science, 206, pp. 563-565.

Hansen, B. L. and C. C. Langway, Jr., 1966: Deep core drilling in ice and core analysis at Camp Century, Greenland, 1961-1966. Antarctic J. U.S., 1, p. 207-208.

Heidam, N. Z., 1981: On the origin of the Arctic aerosol: A statistical approach. Atmos. Env., 15, p. 1421-1427.

Heintzenburg, J., 1980: Measurement of light absorption and elemental carbon in atmospheric aerosol samples from remote locations. Presented at the Int. Symp. Part. Carbon: Atmospheric life cycles, Oct. 12-14, 1980, Gen. Motors. Res. Lab., Warren, Mich., USA.

Heintzenburg, J., 1982: Size-segregated measurements of particulate elemental carbon and aerosol light absorption at remote Arctic locations. Atmos. Evniron., In Press.

Herron, M. M., C. C. Langway Jr., H. V. Weiss and J. H. Cragin, 1977: Atmospheric trace metals and sulphate in the Greenland Ice sheet. Geochim. et Cosmochim. Acta, 41, p. 915-920.

Higuchi, K. and H. Wushiki, 1970: Observations of pre-activated ice nuclei in the atmosphere at subzero temperature. J. Met. Soc. Jap., 48, p. 250-254.

Hindman, E. E., P. V. Hobbs and L. F. Radke 1977: Cloud condensation nuclei from a paper mill. Part I. Measured effects on clouds. J. Appl. Meteor., 16, p. 745-752.

Hobbs, P. V., 1969: Ice multiplication in clouds. J. Atmos. Sci., 26, p. 315-318.

Hobbs, P. V. and L. F. Radke, 1969: Cloud condensation nuclei from a simulated forest fire. Science, 163, p. 279-280.

Hobbs, P. V., L. F. Radke and S. J. Shumway, 1970: Cloud condensation nuclei from industrial sources and their apparent influence on precipitation in Washington state. J. Atmos. Sci., 27, p.81-89.

Hobbs, P. V., J. L. Stith and L. F. Radke, 1980: Cloud-active nuclei from coal-fired electric power plants and their interactions with clouds. J. Appl. Meteor., 19, p. 439-451.

Holmgren, B., 1971: Climate and energy exchange on a sub-polar ice cap in summer. Arctic Institute of North America Devon Island Expedition, 1961-1-63: Madelanden fran Uppsala Universitets Meteorologiska Institutionen, No. 107-112.

Holmgren, B., G. E. Shaw and G. Weller, 1974: Turbidity in the Arctic atmosphere. AIDJEX Bull., No. 27, p. 135-148.

Hopple, W. A., J. E. Dinger and R. E. Ruskin, 1973: Vertical profiles of CCN at various geophysical locations. J. Atmos. Sci., 30, p. 1410-1419.

Huffman, P. J. and G. Vali, 1973: The effect of vapor depletion on ice nucleus measurements with membrane filters. J. Appl. Meteor., 12, 1018-1024.

Isono, K., M. Komabayashi, T. Takeda, T. Tanaka, K. Iwai and M. Fujiwara, 1971: Concentration and nature of ice nuclei in rim of the North Pacific Ocean. Tellus, 23, p.40-59.

Isono, K., M. Komabayshi and A. Ono, 1959: Volcanoes as a source of atmospheric ice nuclei. Nature, 183, pp. 317-318.

Isono, K., M. Komabayashi and A. Ono, 1959: The nature and origin of ice nuclei in the atmosphere. J. Roy. Meteor. Soc. Japan, 37, p. 211-233.

Jayaweera, K. O. L. F. and T. Ohtake, 1973: Concentrations of ice crystals in Arctic stratus clouds. J. Rech. Atmos., 7, p. 199-207.

Jayaweera, K. O. L. F. and P. Flanagan, 1981: Concentration and nature of biogenic ice nuclei over the Arctic Ocean. Prog. and Abs., IAMAP-ICCP, Hamburg, Germ., 17-28 Aug., 1981, p. 35.

Junge, C. E., 1960: Sulphur in the atmosphere. J. Geophys. Res., 65, p. 227-237.

Koide, M. and E. D. Goldberg, 1971: Atmospheric sulphur and fossil fuel combustion. J. Geophys. Res., 76, p. 6589-6596.

Koptev, A. P. and A. I. Voskresenskiiy, 1966: "On the radiation properties of clouds". In: Soviet Data on the Arctic Heat Budget and Its Climatic Influence. ed. by J. O. Fletcher, B. Keller and S. M. Olencoff, Corp. Mem. RM-5003 PR., Santa Monica, CA, 205 pp.

Kumai, M. and K. E. Francis, 1962: Nuclei in snow crystals on the Greenland ice cap under natural and artificially stimulated conditions. J. Atmos. Sci., 19, p. 474-481.

Kumai, M., 1965: The properties of marine and marine fog at Barrow, Alaska. Proc. Int. Conf. Cloud Phys., Meteor. Soc. Japan, p. 52-56.

Kumai, M., 1973: Arctic fog droplet size distribution and its effect on light attenuation. J. Atmos. Sci., 30, p. 635-643.

Laktionov, A. G., 1969: On the distribution of the condensation cloud nuclei in the free atmosphere and the relationship between the concentration of cloud nuclei and the drop concentration in clouds. Akademia Nauk. S. S. S. R., Doklady, Ser. Matem., Fizika, 176, 1/3, p. 315-318.

Magono, C., ed., 1978: Snow crystals in the Arctic Canada, Interim Rep. of "The Scientific Research on the Snow Crystals of Cold Temperature Type in Canada", under Program of Overseas Scientific Survey, Ministry of Education, Hokkaido University, Japan, 172 pp.

Mamane, Y. and R. F. Pueschel, 1980: Formation of sulphate particles in the plume of the Four Corners power plant. J. Appl. Meteor., 19, p. 779-790.

Mason, B. J. and J. Maybank, 1958: Ice-nucleating properties of some natural mineral dusts. Quart. J. Roy. Meteor. Soc., 84, p. 235-241.

Mason, B. J., 1960: Ice-nucleating properties of clay minerals and stony meteorites. Quart. J. Roy. Meteor. Soc., 86, p. 552-556.

Meagher, J. F., L. Stockburger, E. M. Bailey and O. Huff, 1978: The oxidation of sulphur dioxide to sulphate aerosols in the plume of a coal-fired power plant. Atmos. Environ., 12, p. 2197-2203.

Mitchell, M., 1956: Visual range in the polar regions with particular reference to the Alaskan Arctic. J. Atmos. Terr. Phys., Special Supplement, p. 195-211.

Mroz, E. J. and W. H. Zoller, 1975: Composition of atmospheric particulate matter from the eruption of Heimaey, Iceland. Science, 190, p. 461-464.

Murozumi, M., T. J. Chow and C. Patterson, 1969: Chemical concentrations of pollutant lead aerosols, terrestrial dusts and sea salts in Greenland and Antarctic snow strata. Geochim. et Cosmochim. Acta, 33, p. 1247-1294.

Ohtake, T., 1981: A new cloud condensation nuclei counter. Prog. and Abs., IAMAP-ICCP, Hamburg, Germ., 17-28 Aug., 1981, p. 37.

Ottar, B., 1981: The transfer of airborne pollutants to the Arctic region. Atmos. Environ., 15, p. 1439-1445.

Parungo, F. P., H. Ackerman, H. Proulx and R. F. Pueschel, 1978: Nucleation properties of fly ash in a coal-fired power plant plume. Atmos. Environ., 12, p. 920-935.

Patterson, D. E. and R. B. Husar, 1981: A direct simulation of hemispherical transport of pollutants. Atmos. Environ., 15, p. 1479-1482.

Pruppacher, H. R. and J. D. Klett, 1980: Microphysics of Clouds and Precipitation. D. Reidel Pub. Co., Dordrecht, Holland.

Pueschel, R. F., F. P. Parungo, E. W. Barrett, D. L. Wellman and H. Proulx, 1979: Meteorological effects of oil refinery operations in Los Angeles. NOAA Tech. Mem. ERL-APCL-22, 62 pp.

Radke, L. F., P. V. Hobbs and E. Pinnons, 1976: Observations of cloud condensation nuclei, sodium containing particles, ice nuclei and the light-scattering coefficient near Barrow, Alaska. J. Appl. Meteor., 15, p. 982-995.

Rahn, K. A., 1976: The chemical composition of the atmospheric aerosol. University of Rhode Island Technical Report, 265 pp.

Rahn, K. A., R. D. Borys and R. A. Duce, 1976: Tropospheric halogen gases: inorganic and organic components. Science, 192, p. 549-550.

Rahn, K. A., R. D. Borys and G. E. Shaw, 1977: Air pollution in the Arctic: some emerging ideas. Presented at the Conference "Sources and significance of natural and man-made aerosols in the Arctic", Norwegian Institute for Air Research, Lillestrom, Norway, 27-28 April 1977.

Rahn, K. A., R. D. Borys and G. E. Shaw, 1977: The Asian source of Arctic Haze bands. Nature, 268, p. 713-715.

Rahn, K. A., 1979: The Arctic Air Sampling Network, Arctic Bull., 2, p. 342-346.

Rahn, K.A. and R.J. McCaffrey, 1979: Compositional differences between Arctic aerosol and snow. Nature, 280, pp. 479-480.

Rahn, K. A. and R. J. McCaffrey, 1980: On the origin and transport of the winter arctic aerosol. In:Aerosols: Anthropogenic and Natural, Sources and Transport, Ann. N. Y. Acad. Sci., 338, p. 486-503.

Rahn, K.A. and N.Z. Heidam, 1981: Progress in Arctic air chemistry, 1977-1980: A comparison of the first and second symposia. Atmos. Environ., 15, pp. 1345-1348.

Reiter, E. R., 1969: Atmospheric Transport Processes Part I; Energy Transfers and Transformations. NTIS, TID-24868, 253 pp.

Reiter, E. R., 1981: Planetary-wave behavior and Arctic air pollution. Atmos. Environ., 15, p. 1465-1471.

Saxena, V. K., 1981: Sources of cloud condensation nuclei in the Arctic region. Prog. and Abs., IAMAP-ICCP, Hamburg, Germ., 17-28 Aug., 1981, p. 16.

Saxena, V. K. and R. E. Baier, 1981: Evidence of biogenic nuclei involvement in the Anarctic coastal clouds. Prog. and Abs. IAMAP-ICCP, Hamburg, Germ., 17-28 Aug. 1981, p.56.

Schaefer, V. J., 1969: The inadvertant modification of the atmosphere by air pollution. Bull. Amer. Meteor. Soc., 50, p. 199-206.

Schnell, R. C., S. W. Miller and P. A. Allee, 1981: Biogenic ice nuclei: Intercomparison measurements using a thermal diffusion chamber, an NCAR counter and a filter-drop freezer technique. Prog. and Abs. IAMAP-ICCP, Hamburg, Germ., 17-28 Aug., 1981, p. 30.

Shaw, G. E. and G. Wendler, 1972: Atmospheric turbidity measurements at McCall Glacier in Northeast Alaska. Proceedings of the Atmospheric Radiation Conference, Fort Collins, Colorado, August 7-9, 1972.

Shaw, G. E., 1975: The vertical distribution of tropospheric aerosols at Barrow, Alaska. Tellus, 27, p. 39-49.

Shaw, G. E., 1981: Eddy diffusion transport of Arctic pollution from the mid-latitudes: a preliminary model. Atmos. Env., 15, p. 1483-1490.

Squires, P., 1966: An estimate of the anthropogenic production of cloud nuclei. J. Rech. Atmos., 2., pp. 297-307.

Stevenson, C. M., 1968: An improved Millpore filter technique for measuring the concentrations of freezing nuclei in the atmosphere. Quart. J. Roy. Met. Soc., 94, 35-43.

Tabatabai, M. A., 1974: A rapid method for determination of sulphate in water samples. Envir. Lett. 7, 237-243.

Twomey, S., 1959: The nuclei of natural cloud formation II. The supersaturation in natural clouds and the variation of cloud droplet concentrations. Geofis. Pura Appl., 43, p. 243-249.

Twomey, S., 1963: Measurements of natural cloud nuclei. J. Rech. Atmos., 1, p. 101-105.

Twomey, S. and G.T. Severynse, 1964: On the relationship between sizes of particles and their ability to nucleate condensation of natural clouds. J. Rech. Atmos., 1, p. 81-85.

Twomey, S. and T. A. Wojciechowski, 1969: Observations of the geographical variation of cloud nuclei. J. Atmos. Sci., 26, p. 684-688.

Twomey, S., 1977: The influence of pollution on the short wave albedo of clouds. J. Atmos. Sci., 34, p. 1149-1152.

Van Valin, C. C., R. F. Pueschel and F. P. Parungo, 1975: Formation of cloud and ice nuclei by the combustion of crude oil. J. Appl. Meteor., 14, p. 1200-1203.

Van Valin, C. C., R. F. Pueschel, F. P. Parungo and R. A. Proulx, 1976: Cloud and ice nuclei from human activities. Atmos. Environ., 10, p. 27-31.

Van Valin, C. C. and R. F. Pueschel, 1976: Cloud nuclei formation in the plume of a coal-fired power plant. T. Amer. Geophys. Union, 57, p. 925.

Weickmann, H. K., 1972: Man-made weather patterns in the Great Lakes Basin. Weatherwise, 25, p. 260-268.

Welch, R. and S. Cox, 1980: Solar radiation and clouds. Met. Mono., 17, No. 39, 96 pp.

Weschler, C. J., 1981: Identification of selected organics in the Arctic aerosol. Atmos. Env., 15, p. 1365-1369.

Winchester, J. W. and R. A. Duce, 1966: Coherence of iodine and bromine in the atmosphere of Hawaii, northern Alaska and Massachusetts. Tellus, 18, p. 287-291.

Witte, H. J., 1968: Airborne observations of cloud particles and infrared flux density (8-14 μm) in the Arctic. Masters Thesis, Dept. Atmos. Sci., Univ. of Wash., Seattle, WA 98195, 102 pp.

Zoller, W. H., G. E. Gordon, E. S. Gladney and A. G. Jones, 1973: The sources and distribution of vanadium in the atmosphere: Trace Elements in the Environment, Advances in Chemistry Series, No. 123, American Chemical Society, Washington, D.C., p. 31-47.

APPENDIX A

Table 1

Aerosol composition (ng/m^3) and 1σ uncertainties, Narragansett, Rhode Island, USA, summer 1979.

Sample	1	2	3	4	5	6	7
Dates (June)	12-13	13-14	14-15	15-16	16-17	17-18	18-19
Al	320 ± 7	290 ± 7	490 ± 10	930 ± 20	850 ± 20	320 ± 10	360 ± 8
Ba	3.3 ± 0.7	5.3 ± 0.9	5.8 ± 0.9	9.0 ± 1.1	11.1 ± 0.4	2.7 ± 0.2	1.2 ± 0.1
Br	9.9 ± 0.4	26 ± 1	39 ± 2	72 ± 3	43 ± 2	13.9 ± 0.9	32.4 ± 1.2
Ca	200 ± 20	240 ± 40	350 ± 80	490 ± 120	<200	150 ± 70	<90
Cl	118 ± 14	150 ± 15	360 ± 30	250 ± 20	850 ± 50	740 ± 50	510 ± 40
Cu	<4	1.63 ± 0.20	2.9 ± 0.7	23 ± 9	1.33 ± 0.30	<5	2.7 ± 0.4
Dy	0.036 ± 0.003	0.044 ± 0.003	0.066 ± 0.005	0.111 ± 0.007	0.084 ± 0.004	0.027 ± 0.004	0.051 ± 0.005
I	0.49 ± 0.09	0.90 ± 0.10	3.8 ± 0.2	5.1 ± 0.2	5.3 ± 0.3	3.4 ± 0.2	1.60 ± 0.12
In	0.033 ± 0.002	0.0195 ± 0.0025	0.022 ± 0.003	0.012 ± 0.003	0.012 ± 0.001	0.0039 ± 0.004	0.0084 ± 0.0018
Mg	<4	140 ± 10	<380	420 ± 30	63 ± 9	320 ± 30	20 ± 5
Mn	6.5 ± 0.2	8.3 ± 0.3	10.0 ± 0.40	23 ± 1	14.4 ± 0.5	5.8 ± 0.2	5.6 ± 0.2
Na	99 ± 5	151 ± 6	640 ± 20	1000 ± 30	1950 ± 50	940 ± 30	670 ± 20
Sn	2.4 ± 0.4	7.0 ± 0.6	<20	43 ± 3	10 ± 2	<15	<15
Ti	24 ± 4	30 ± 4	37 ± 8	82 ± 10	33 ± 5	23 ± 2	34 ± 5
V	1.9 ± 0.7	6.4 ± 0.2	15.4 ± 0.40	33 ± 1	21 ± 2	9.4 ± 0.3	12.0 ± 0.4
SO_4	2440	3210	5000	14570	12630	4330	10860

Table 2

Aerosol composition (ng/m^3) and 1σ uncertainty, Narragansett, Rhode Island, USA, winter 1980.

Sample	1	2	3	4	5	6	7	8	9	10
Dates (Jan)	6-7	7-8	8-9	9-10	10-11	11-12	12-13	13-14	14-15	15-16
Al	197 ± 9	167 ± 9	500 ± 10	210 ± 10	230 ± 10	39 ± 4	590 ± 20	190 + 5	90 ± 5	170 ± 5
Ba	2.5 ± 0.9	<1	85 ± 10	67 ± 11	<2	28 ± 2	54 ± 7	30 ± 12	<10	<8
Br	35 ± 2	55 ± 3	50 ± 2	21 ± 1	41 ± 2	65 ± 4	13.5 ± 0.3	57 ± 3	70 ± 4	32 ± 2
Ca	12 ± 8	340 ± 170	420 ± 90	170 ± 50	130 + 60	<340	490 ± 60	200 ± 40	<290	<160
Cl	4600 ± 300	4400 ± 60	470 ± 50	350 ± 40	1400 ± 100	10960 ± 100	147 ± 25	2900 ± 200	9500 ± 100	5000 ± 400
Cu	<7	17 ± 8	4.7 ± 0.8	<6	3.6 ± 0.7	<0.6	4.6 ± 0.9	2.9 ± 0.8	<15	<10
Dy	NA	NA	NA	NA	0.0401 ± 0.0018	<0.04	0.063 ± 0.003	0.028 + 0.002	<0.05	0.0124 ± 0.0015
I	1.94 ± 0.35	5.0 ± 0.6	1.49 ± 0.15	1.91 ± 0.13	2.5 ± 0.2	2.8 ± 0.7	1.0 ± 0.1	3.0 ± 0.3	6.1 ± 0.8	1.07 ± 0.30
In	<0.005	0.025 ± 0.002	0.0193 ± 0.0040	<0.004	0.0045 ± 0.0005	<0.002	0.0158 ± 0.0020	<0.008	<0.02	<0.02
Mg	330 ± 30	750 ± 190	490 ± 90	240 ± 100	300 ± 70	690 ± 180	370 ± 90	<70	740 ± 180	760 ± 140
Mn	3.4 ± 0.2	8.5 ± 0.3	12.8 ± 0.5	13.9 + 0.5	7.9 ± 0.3	5.9 ± 0.4	11.6 ± 0.4	5.2 ± 0.2	7.6 ± 0.3	5.4 ± 0.2
Na	3400 ± 60	3900 ± 70	340 ± 10	260 ± 10	1150 ± 30	7100 ± 200	165 ± 10	2100 ± 50	6700 ± 400	3200 ± 70
Sn	4.6 ± 0.9	6.4 ± 0.4	17 ± 2	8.9 ± 1.2	<20	13 ± 2	<13	26 ± 2	11 ± 2	<30
Ti	3.3 ± 0.6	34 ±]	24 ± 5	21 ± 2	22 ± 5	<20	56 ± 7	2.4 ± 0.5	15 ± 2	<20
V	23 ± 1.0	15.5 ± 0.5	11.6 ± 0.4	10.6 ± 0.3	21 ± 2.0	2.4 ± 0.2	7.2 ± 0.1	28 ± 1	116 ± 4	36 ± 1
SO_4	7740	10900	9860	9410	9350	7550	6050	7690	7780	8030

NA: Data not available

Table 3

Aerosol composition (ng/m^3) and 1σ uncertainty, Barrow, Alaska, USA, summer, 1979.

Sample	1	2	3	4	5	6	7	8	9	10
Dates (July-Aug)	27 - 29	30 - 31	31 - 1	1 - 2	2 - 3	3 - 4	4 - 5	5 - 6	6 - 7	7 - 8
Al	11.9 ± 0.4	6.0 ± 0.5	6.4 ± 0.5	4.8 ± 0.5	102 ± 3	9.3 ± 0.5	2.3 ± 0.3	4.4 ± 0.5	3.1 ± 0.4	3.3 ± 0.5
Ba	0.135 ± 0.018	< 2	1.27 ± 0.56	< 3	4.8 ± 0.2	< 2	1.19 ± 0.08	2.3 ± 0.2	2.5 ± 0.2	< 1
Br	3.7 ± 0.3	2.4 ± 0.4	1.45 ± 0.47	1.65 ± 0.48	2.2 ± 0.4	1.05 ± 0.36	1.60 ± 0.29	2.7 ± 0.4	2.5 ± 0.2	0.68 ± 0.48
Ca	50	< 40	< 15	< 50	< 70	< 40	< 30	< 30	< 80	< 10
Cl	440 ± 30	670 ± 45	185 ± 17	1270 ± 80	1170 ± 70	830 ± 50	540 ± 35	920 ± 60	780 ± 50	220 ± 20
Cu	< 3	< 0.5	< 0.6	< 4	< 7	< 1	2.7 ± 0.4	< 7	< 6	< 4
Dy	< 0.008	< 0.0004	< 0.003	< 0.006	0.0092 ± 0.0006	0.0043 ± 0.0004	0.003	< 0.01	< 0.005	< 0.003
I	1.51 ± 0.06	2.0 ± 0.1	3.0 ± 0.1	3.9 ± 0.1	2.6 ± 0.1	2.0 ± 0.1	1.23 ± 0.07	0.84 ± 0.08	1.76 ± 0.09	1.75 ± 0.08
In	< 0.002	0.00071 ± 0.00013	< 0.003	0.0031 ± 0.0003	< 0.005	0.00046±0.00011	0.00142±0.00023	<0.004	< 0.004	< 0.002
Mg	58 ± 13	63 ±27	27 ± 4	< 60	57 ± 7	13 ± 2	49 ± 15	93 ± 30	< 40	< 30
Mn	0.46 ± 0.02	0.24 ± 0.03	0.141 ± 0.014	0.150±0.023	0.81 ± 0.04	0.081 ± 0.012	0.050±0.010	0.0129±0.0046	0.061±0.018	< 0.09
Na	270 ± 7	400 ± 11	90 ± 5	108 ± 6	220 ± 7	26 ± 3	310 ± 10	560 ± 15	470 ± 15	110 ± 5
Sn	NA	NA	NA	NA	5.6 ± 0.8	2.6 ± 0.5	---	---	---	---
Ti	< 2.5	< 0.5	< 1.1	3.3 ± 2.0	7.3 ± 2.0	<2	<2	9.1 ± 0.9	4.1 ± 0.8	<0.6
V	0.0127 ± 0.0058	< 0.009	0.0191 ± 0.0070	<0.04	0.195 ± 0.018	0.032 ± 0.003	< 0.02	< 0.03	< 0.03	< 0.02
SO_4	278	< 30	81	76	< 40	6 ± 6	106	46 ± 6	48 ± 7	75

NA - Data not available

Table 4

Aerosol composition (ng/m^3) and 1σ uncertainty, Barrow, Alaska, USA, winter 1980.

Sample	1	2	3	4	5	6	7	8	9	10
Dates (Mar-April)	24-25	25-26	26-27	27-28	28-29	29-30	30-31	31-1	1-2	2-3
Al	21 ± 0.8	21 ± 0.8	22 ± 0.8	19 ± 0.8	19.7 ± 0.9	21 ± 0.9	21 ± 0.9	18.8 ± 0.9	41 ± 1.0	31 ± 1.0
Ba	<5	<2	<3	<2	<2	<3	<3	<3	<7	<3
Br	62 ± 6	43 ± 2	69 ± 2	83 ± 3	70 ± 3	78 ± 3	91 ± 3	61 ± 2	72 ± 2	37 ± 2
Ca	33 ± 6	<30	<90	20 ± 6	<40	<30	47 ± 16	86 ± 10	140 ± 30	49 ± 20
Cl	330 ± 30	230 ± 20	380 ± 30	290 ± 30	400 ± 30	400 ± 30	520 ± 40	1160 ± 80	6100 ± 400	1270 ± 90
Cu	1.1 ± 0.4	<1	<2	1.9 ± 0.4	0.95 ± 0.30	<1	1.2 ± 0.7	<0.2	1.5 ± 0.5	<2
Dy	<0.005	0.0018 ± 0.0008	<0.009	<0.002	<0.005	0.0078±0.0020	0.0140±0.0015	<0.001	<0.007	<0.002
I	2.2 ± 0.1	1.29 ± 0.10	1.59 ± 0.10	2.1 ± 0.10	1.94 ± 0.10	2.5 ± 0.10	2.9 ± 0.20	1.94 ± 0.20	1.22 ± 0.40	1.04 ± 0.13
In	<0.005	0.004±0.0016	0.0006±0.0003	0.00049±0.00019	<0.009	<0.0005	<0.0005	<0.01	0.026 ± 0.002	<0.0002
Mg	76 ± 20	56 ± 15	53 ± 15	74 ± 15	97 ± 20	88 ± 15	47 ± 15	15 ± 3	580 ± 40	140 ± 30
Mn	1.05 ± 0.06	1.08 ± 0.06	1.32 ± 0.06	1.22 ± 0.06	1.11 ± 0.06	1.44 ± 0.07	1.09 ± 0.005	0.94 ± 0.05	0.98 ± 0.09	0.93 ± 0.05
Na	260 ± 10	190 ± 10	260 ± 10	220 ± 10	310 ± 10	340 ± 10	380 ± 10	660 ± 20	3100 ± 60	720 ± 20
Sn	12.5 ± 0.8	7.5 ± 2.0	7.8 ± 0.7	10.3 ± 0.7	6.9 ± 0.6	7.7 ± 0.7	4.7 ± 0.6	<0.6	14 ± 2.0	11.4 ± 0.8
Ti	3.2 ± 0.4	0.7 ± 0.2	3.4 ± 1.5	<2	<2	4.6 ± 2.0	<3	0.7 ± 0.2	<10	8.0 ± 3.0
V	0.87 ± 0.04	0.89 ± 0.04	1.18 ± 0.05	1.00 ± 0.05	0.93 ± 0.04	1.19 ± 0.05	0.82 ± 0.04	0.44 ± 0.03	0.36 ± 0.03	0.36 ± 0.03
SO_4	2440	3690	3540	3800	4190	4760	1960	2450	3110	2020

Table 5

Aerosol Composition (ng/m^3) and 1σ uncertainty, Poker Flat, Alaska, USA, summer, 1979

Sample	1	2	3	4	5	6	7	8	9
Dates (July)	17 - 18	18 - 19	19 - 20	20 - 21	21 - 22	22 - 23	23 - 24	24 - 25	25
Al	210 ± 5	197 ± 5	380 ± 10	210 ± 5	56 ± 8	36 ± 1	290 ± 7	420 ± 9	25 ± 1
Ba	2.1 ± 0.4	10.6 ± 2.3	7.2 ± 1.0	2.2 ± 0.3	1.28 ± 0.09	4.2 ± 0.2	5.0 ± 1.6	5.7 ± 0.5	2.9 ± 1.2
Br	2.8 ± 0.5	3.5 ± 0.4	4.1 ± 0.6	0.75 ± 0.19	1.89 ± 0.17	3.9 ± 0.3	3.3 ± 0.4	1.25 ± 0.40	1.50 ± 0.24
Ca	< 40	41 ± 10	< 70	< 40	< 2000	< 60	123 ± 24	95 ± 23	56 ± 10
Cl	< 60	44 ± 15	75 ± 17	53 ± 4	85 ± 6	350 ± 20	64 ± 5	79 ± 5	640 ± 40
Cu	3.2 ± 0.4	< 0.4	< 0.7	< 5	< 1	< 1	< 2	< 1	< 3
Dy	NA	NA	NA	NA	NA	NA	NA	NA	NA
I	1.11 ± 0.11	1.51 ± 0.10	2.1 ± 0.1	0.16 ± 0.04	0.36 ± 0.04	< 0.06	0.16 ± 0.02	0.16 ± 0.02	0.4 ± 0.1
In	< 0.006	0.00035 ± 0.00012	< 0.001	< 0.002	< 0.0009	0.0089 ± 0.0005	< 0.002	< 0.002	< 0.003
Mg	72 ± 9	< 50	105 ± 9	93 ± 30	24 ± 7	< 50	108 ± 8	280 ± 70	19 ± 6
Mn	1.76 ± 0.07	2.1 ± 0.1	3.7 ± 0.1	1.70 ± 0.06	1.36 ± 0.05	1.09 ± 0.04	5.4 ± 0.2	4.4 ± 0.4	0.70 ± 0.05
Na	30 ± 5	74 ± 4	121 ± 6	28 ± 3	9.6 ± 2.1	11.5 ± 2.4	76 ± 4	72 ± 3	4.4 ± 5.1
Sn	1.4 ± 0.5	2.6 ± 0.5	11 ± 2	< 15	< 0.3	1.7 ± 0.3	< 4	< 7	< 4
Ti	11 ± 1	24 ± 4	17 ± 3	9 ± 2	< 3	2.1 ± 0.6	270 ± 3	27 ± 4	5 ± 2
V	0.29 ± 0.03	0.30 ± 0.03	0.74 ± 0.05	0.39 ± 0.02	0.15 ± 0.01	0.077 ± 0.009	<0.07	0.72 ± 0.04	0.047 ± 0.007
SO_4	NA	NA	NA	335	116	436	174	91	< 50

NA - Data Not Available

Table 6

Aerosol composition (ng/m^3) and 1σ uncertainty, Poker Flat, Alaska, USA, winter 1980.

Sample	1	2	3	4	5	6	7	8	9	10
Dates (April)	4-5	5-6	6-7	7-8	8-9	9-10	10-11	11-12	12-13	13-14
Al	210 ± 10	141 ± 5	95 ± 3	250 ± 10	140 ± 5	300 ± 10	94 ± 5	125 ± 5	86 ± 5	310 ± 8
Ba	3.2 ± 0.5	1.8 ± 0.6	1.9 ± 0.6	2.8 ± 0.6	0.74 ± 0.6	5.1 ± 0.5	1.42 ± 0.5	8.2 ± 0.8	4.7 ± 0.7	13.9 ± 1.5
Br	6.8 ± 0.4	7.1 ± 0.4	6.9 ± 0.4	6.5 ± 0.4	7.5 ± 0.4	11.4 ± 0.6	27 ± 1	6.9 ± 0.4	4.4 ± 0.4	<2
Ca	65 ± 15	<20	13 ± 4	53 ± 15	35 ± 10	<15	73 ± 9	32 ± 10	<20	79 ± 20
Cl	<50	<20	<50	<60	<20	102 ± 30	101 ± 20	87 ± 25	46 ± 20	<20
Cu	<2	<2	2.3 ± 0.8	<0.9	1.4 ± 0.6	2.3 ± 0.3	1.8 ± 0.2	<3	<0.6	<0.4
Dy	<0.02	<0.009	0.038 ± 0.018	<0.02	<0.01	<0.02	<0.006	0.125 ± 0.010	0.081 ± 0.018	0.062 ± 0.015
I	0.87 ± 0.06	1.03 ± 0.06	1.12 ± 0.06	0.81 ± 0.06	0.99 ± 0.07	0.95 ± 0.04	0.98 ± 0.04	1.14 ± 0.07	1.05 ± 0.06	0.77 ± 0.12
In	<0.0005	<0.0005	<0.001	<0.002	<0.005	<0.003	<0.002	<0.003	<0.001	<.003
Mg	110 ± 30	56 ± 15	69 ± 20	47 ± 20	<60	75 ± 25	36 ± 15	<80	<50	<80
Mn	1.88 ± 0.07	1.40 ± 0.07	1.06 ± 0.06	2.5 ± 0.09	1.75 ± 0.08	3.3 ± 0.20	1.93 ± 0.08	4.7 ± 0.2	3.3 ± 0.2	13.8 ± 0.5
Na	81 ± 5	83 ± 4	87 ± 4	99 ± 4	84 ± 4	146 ± 5	181 ± 6	200 ± 7	127 ± 6	230 ± 8
Sn	<0.7	1.2 ± 0.3	<0.1	6.3 ± 0.5	3.1 ± 0.4	<5	<4	<3	<2	<5
Tl	11 ± 2	8.3 ± 1.3	4.5 ± 1.0	9.9 ± 1.5	7.8 ± 1.5	12 ± 2.5	7.3 ± 0.9	2.9 ± 0.8	3.3 ± 0.9	16 ± 7
V	0.43 ± 0.03	0.40 ± 0.03	0.36 ± 0.02	0.61 ± 0.04	0.34 ± 0.03	0.64 ± 0.04	0.48 ± 0.02	0.26 ± 0.02	0.18 ± 0.02	0.50 ± 0.04
SO_4	2160	1030	1400	1230	1040	1140	1720	2550	2570	1930

Table 7

Aerosol composition (ng/m^3) and 1σ uncertainty, Pangnirtung, N.W.T., Canada, summer, 1979.

Sample	1	2	3	4	5	6	7	8	9
Dates (June-July)	22-23	23-24	24-25	25-26	26-27	27-28	28-29	29-30	30-1
Al	22 ± 2	5 ± 1	< 20	23 ± 2	340 ± 8	292 ± 7	214 ± 5	40 ± 2	22 ± 2
Ba	< 1	< 0.3	< 1	< 1	5 ± 1	4 ± 1	3 ± 1	< 1	< 1
Br	< 0.9	1.7 ± 0.3	< 0.8	0.9 ± 0.4	3.8 ± 0.5	1.9 ± 0.4	< 2	1.9 ± 0.5	< 1
Ca	< 40	< 40	< 10	< 40	88 ± 32	180 ± 34	82 ± 33	< 90	< 30
Cl	135 ± 22	44 ± 15	360 ± 35	68 ± 19	78 ± 17	172 ± 21	240 ± 25	270 ± 27	34 ± 16
Cu	< 0.1	< 0.1	< 1	< 1	< 2	2.9 ± 0.4	< 2	< 1	< 1
Dy	< 0.004	< 0.003	< 0.005	< 0.004	0.030 ± 0.003	0.031 ± 0.003	0.030 ± 0.003	0.0022 ± 0.0003	<0.003
I	0.94 ± 0.13	0.33 ± 0.10	1.19 ± 0.15	1.57 ± 0.13	2.5 ± 0.1	0.99 ± 0.12	1.03 ± 0.12	1.18 ± 0.13	1.79 ± 0.13
In	< 0.009	< 0.003	< 0.007	< 0.005	0.009 ± 0.002	0.006 ± 0.002	< 0.006	< 0.0006	< 0.003
Mg	< 10	< 20	< 20	9 ± 4	116 ± 30	121 ± 55	19 ± 5	< 60	< 10
Mn	0.21 ± 0.03	0.085 ± 0.024	0.139± 0.036	0.29 ± 0.032	3.4 ± 0.1	2.5 ± 0.1	1.69 ± 0.07	0.61 ± 0.04	0.119 ± 0.026
Na	91 ± 5	52 ± 3	230 ± 8	60 ± 4	145 ± 5	220 ± 7	220 ± 7	194 ± 7	17.6 ± 2.9
Sn	---	---	---	---	---	---	---	---	---
Ti	< 2	< 1	< 1	< 2	21 ± 5	15.9 ± 2.4	11.1 ± 2.5	< 1	< 1
V	< 0.02	< 0.02	0.02	0.048 ± 0.018	0.47 ± 0.04	0.36 ± 0.03	0.170 ± 0.023	0.093 ± 0.021	0.038±0.015
SO_4	NO SULPHATE DATA AVAILABLE								

Table 8

Aerosol composition (ng/m^3) and 1σ uncertainty, Pangnirtung, N.W.T., Canada, winter 1980.

Sample	1	2	3	4	5	6	7
Dates (Jan-Feb)	26-27	27-28	28-29	29-30	30-1	1-3	3-4
Al	194 ± 25	440 ± 50	230 ± 30	240 ± 30	86 ± 10	77 ± 10	88 ± 10
Ba	2.7 ± 0.7	5.7 ± 0.5	2.5 ± 1.1	4.9 ± 1.7	0.9 ± 0.4	5.4 ± 1.2	1.23 ± 0.20
Br	5.8 ± 0.4	4.4 ± 0.10	5.0 ± 0.6	9.9 ± 0.7	2.4 ± 0.3	14.2 ± 0.70	15.6 ± 0.6
Ca	60 ± 25	176 ± 30	83 ± 35	< 70	33 ± 15	< 60	< 15
Cl	210 ± 25	520 ± 50	480 ± 50	580 ± 50	129 ± 20	165 ± 34	50 ± 20
Cu	< 2	< 0.8	< 0.7	1.3 ± 0.3	< 1	< 2	< 1
Dy	0.093 ± 0.009	0.0113 ± 0.0046	< 0.009	< 0.008	0.0194 ± 0.0052	< 0.003	< 0.004
I	1.14 ± 0.09	2.4 ± 0.2	4.2 ± 0.3	3.4 ± 0.2	1.51 ± 0.08	2.3 ± 0.2	1.70 ± 0.09
In	< 0.004	0.0104 ± 0.0009	0.0023 ± 0.0007	0.0081 ± 0.008	< 0.002	< 0.007	< 0.004
Mg	58 ± 25	131 ± 15	111 ± 15	84 ± 35	< 20	38 ± 10	< 60
Mn	1.68 ± 0.07	3.5 ± 0.2	1.85 ± 0.10	2.3 ± 0.1	0.56 ± 0.03	0.47 ± 0.05	0.52 ± 0.003
Na	175 ± 6	500 ± 20	390 ± 15	500 ± 15	102 ± 5	165 ± 10	90 ± 5
Sn	< 6	< 10	< 9	11 ± 2	< 4	< 7	< 4
Ti	5.7 ± 1.5	26 ± 6	12 ± 4	6 ± 2	99 ± 20	8 ± 2	< 1
V	0.25 ± 0.03	0.44 ± 0.05	0.31 ± 0.04	0.61 ± 0.05	0.155 ± 0.010	0.21 ± 0.03	0.188 ± 0.020
SO_4	460	< 550	3080	1109 ± 720	180	1240	710

Table 9

Aerosol composition (ng/m^3) and 1σ uncertainty, Iraföss, Iceland, summer, 1979.

Sample	1	2	3	4	5	6	7	8	9
Dates	(Aug.) 17 - 18	18 - 19	19 - 20	20 - 21	21 - 22	22 - 23	23 - 24	24 - 25	25 - 26
Al	346 ± 20	97 ± 8	38 ± 5	168 ± 10	155 ± 10	807 ± 50	752 ± 40	126 ± 9	1196 ± 60
Ba	<2	<1	1.04 ± 0.20	<1	0.35 ± 0.10	<2	3.2 ± 0.3	<0.2	4.2 ± 0.3
Br	5.9 ± 0.7	4.7 ± 0.3	3.0 ± 0.2	1.43 ± 0.25	2.6 ± 0.3	4.1 ± 1.4	8.5 ± 1.7	3.1 ± 0.1	8.3 ± 1.5
Ca	360 ± 50	<50	<60	130 ± 25	85 ± 20	390 ± 80	560 ± 140	120 ± 40	860 ± 130
Cl	670 ± 50	370 ± 25	270 ± 25	88 ± 11	113 ± 11	43 ± 18	2880 ± 120	670 ± 50	350 ± 30
Cu	0.46 ± 0.10	0.22 ± 0.08	1.08 ± 0.15	<1	0.39 ± 0.10	< 6	31 ± 15	< 0.1	12 ± 2
Dy	0.029 ± 0.005	0.0068 ± 0.0006	<0.004	0.0114 ± 0.0020	0.0159 ± 0.0030	0.090 ± 0.015	0.060 ± 0.010	<0.005	0.106 ± 0.010
I	0.97 ± 0.12	0.83 ± 0.12	0.77 ± 0.09	0.70 ± 0.06	0.51 ± 0.05	0.84 ± 0.12	2.0 ± 0.3	1.03 ± 0.21	1.91 ± 0.15
In	<0.003	<0.002	<0.002	0.0028 ± 0.0004	<0.002	0.0019 ± 0.0004	0.0038 ± 0.0005	0.0024 ± 0.0004	<0.005
Mg	117 ± 8	<170	45 ± 5	163 ± 30	51 ± 5	300 ± 30	810 ± 130	154 ± 9	680 ± 30
Mn	8.37 ± 0.60	2.68 ± 0.20	0.81 ± 0.15	3.96 ± 0.30	3.09 ± 0.20	16.7 ± 1.0	22.2 ± 1.4	3.02 ± 0.20	26.6 ± 1.5
Na	490 ± 15	290 ± 10	200 ± 7	90 ± 6	110 ± 6	210 ± 30	2020 ± 70	520 ± 20	525 ± 20
Sn	---	---	---	< 4	---	< 20	43 ± 4	18 ± 5	36 ± 3
Ti	57 ± 5	19.2 ± 0.8	6.5 ± 1.8	24 ± 3	17.1 ± 1.8	142 ± 9	181 ± 3	18 ± 2	188 ± 9
V	1.22 ± 0.09	0.34 ± 0.03	0.15 ± 0.02	0.64 ± 0.06	0.52 ± 0.05	3.0 ± 0.2	4.0 ± 0.3	0.74 ± 0.06	4.8 ± 0.3
SO_4	288	107	298	226	176	512	1810	760	1070

Table 10

Aerosol composition (ng/m^3) and 1σ uncertainty, Irafðss, Iceland, winter 1980.

Sample	1	2	3	4	5	6	7
Dates (Feb)	16-17	17-18	18-19	19-20	20-21	21-22	22-23
Al	8.4 ± 0.7	13.5 ± 0.7	19.8 ± 0.7	90 ± 3	31 ± 4	9.8 ± 2.0	40 ± 2
Ba	5.4 ± 0.6	<4	0.45 ± 0.06	<10	29 ± 2	1.92 ± 0.25	<5
Br	8.6 ± 0.6	5.6 ± 0.5	2.9 ± 0.3	8.9 ± 0.7	7.7 ± 0.6	8.7 ± 0.7	14.1 ± 0.9
Ca	<100	86 ± 10	<15	100 ± 45	240 ± 40	120 ± 60	210 ± 70
Cl	2300 ± 150	1900 ± 120	690 ± 50	3700 ± 50	14200 ± 150	1190 ± 80	2970 ± 190
Cu	<2	<3	<1	2.3 ± 0.4	<2	7.1 ± 0.7	6.9 ± 3.5
Dy	0.0113 ± 0.0012	0.0133 ± 0.0013	<0.002	<0.002	0.0036 ± 0.0010	<0.006	0.0091 ± 0.0011
I	0.33 ± 0.11	0.73 ± 0.12	0.78 ± 0.07	1.20 ± 0.30	2.2 ± 0.9	1.30 ± 0.45	0.30 ± 0.04
In	0.027 ± 0.003	0.0028 ± 0.0013	<0.0002	0.0042 ± 0.0005	0.021 ± 0.009	0.00198 ± 0.00025	<0.01
Mg	120 ± 7	175 ± 40	50 ± 20	112 ± 30	<50	110 ± 9	180 ± 50
Mn	0.34 ± 0.04	0.52 ± 0.02	0.60 ± 0.03	3.9 ± 0.2	1.08 ± 0.20	5.3 ± 0.1	1.52 ± 0.08
Na	1470 ± 40	1210 ± 30	420 ± 15	2150 ± 50	8310 ± 200	515 ± 20	1810 ± 50
Sn	1.2 ± 0.3	4.7 ± 0.5	<3	<4	<0.4	<7	<0.2
Ti	<3	<8	5 ± 2	9 ± 2	<5	<3	5.1 ± 0.8
V	0.051 ± 0.020	0.043 ± 0.006	0.081 ± 0.0.015	0.47 ± 0.06	0.058 ± 0.020	<0.05	0.177 ± 0.040
SO_4	380	280	110	330	710	130	640

Table 11

Aerosol composition (ng/m^3) and 1σ uncertainty, YMER - 80, summer, 1980.

Sample	1	2	3	4	5	6	7	8	9	10	11
Dates (Aug-Sept)	11-12	12-15	15-16	16-18	18-20	20-21	22-23	24-27	27-29	30-31	31-3
Al	250 ± 220	4.2 ± 0.5	4.2 ± 0.6	3.5 ± 0.6	< 0.3	< 0.3	16.7 ± 1.0	30 ± 1.0	3.8 ± 0.5	18.6 ± 0.8	< 0.3
Ba	< 25	< 1.0	1.2 ± 0.1	< 1.0	< 3.0	<4	1.3 ± 0.2	1.6 ± 0.2	0.6 ± 0.4	< 0.6	0.2 ± 0.1
Br	73 ± 5	2.8 ± 0.2	2.2 ± 0.2	3.4 ± 0.3	2.5 ± 0.3	16.7 ± 0.9	8.8 ± 0.6	4.5 ± 0.4	1.53 ± 0.20	0.69 ± 0.20	1.10 ± 0.20
Ca	180 ± 40	1700 ± 700	< 50	< 40	230 ± 40	220 ± 40	22 ± 10	< 30	< 20	< 50	< 20
Cl	40200 ± 1500	720 ± 60	200 ± 30	860 ± 70	2000 ± 150	3600 ± 300	1200 ± 100	900 ± 80	180 ± 30	35 ± 20	180 ± 30
Cu	6.8 ± 1.0	0.077±0.011	1.85 ± 0.51	6.4 ± 0.7	< 4	< 4	< 2	< 1	< 2	1.2 ± 0.4	< 2
Dy	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
I	3.5 ± 1.4	1.26 ± 0.07	1.12±0.07	0.66±0.09	1.68±0.16	1.15 ± 0.24	0.86 ± 0.12	2.1 ± 0.1	2.2 ± 0.1	0.91 ± 0.06	2.1 ± 0.1
In	0.062 ± 0.003	< 0.002	< 0.006	< 0.002	< 0.004	< 0.03	< 0.003	0.0041 ± 0.0003	< 0.006	< 0.001	< 0.002
Mg	350 ± 30	34 ± 15	43 ± 15	70 ± 10	160 ± 20	240 ± 70	170 ± 40	80 ± 25	< 50	15 ± 5	35 ± 5
Mn	1.25 ± 0.06	0.30 ± 0.03	1.37 ± 0.06	0.57±0.04	0.28 ± 0.02	1.42±0.28	< 0.04	0.99 ± 0.06	0.25 ± 0.03	0.32 ± 0.03	0.26±0.03
Na	13700 ± 300	450 ± 20	189 ± 8	540 ± 20	1930 ± 50	2200±60	1550 ± 50	580 ± 20	140 ± 10	64 ± 5	120 ± 10
Sn	< 30	1.1 ± 0.3	1.8 ± 0.3	5.1 ± 0.5	26 ± 2.0	< 20	9.2 ± 0.7	1.5 ± 0.3	< 2	< 3	1.2 ± 0.3
Ti	5.0 ± 1.0	< 3	< 4	< 4	< 4	7 ± 5	< 3	< 3	< 2	< 4	< 4
V	2.6 ± 0.3	0.037 ± 0.011	0.187±0.012	< 0.004	< 0.005	< 0.005	< 0.004	0.031 ± 0.010	< 0.004	< 0.005	< 0.005
SO_4	3680	590	1130	24	490	480	380	230	70	< 160	< 160

Table 12

Aerosol composition (ng/m^3) and 1σ uncertainty, Ny Ålesund, Spitzbergen, Norway, Winter, 1980

Sample	1*	2*	3	4	5*	6	7	8	9	10	11
Dates: Feb-Mar	29-1	1-2	2-3	3-4	4-5	5-6	6-7	7-8	8-9	9-10	10-11
Al	37 ± 2	105 ± 3	71 ± 2	34 ± 1	27 ± 1	66 ± 2	68 ± 2	55 ± 2	300 ± 8	25 ± 1	36 ± 1
Ba	1.68 ± 0.90	2.4 ± 0.4	1.54 ± 0.30	<0.7	<1	5.9 ± 0.6	2.7 ± 0.8	2.2 ± 0.6	< 2	1.3 ± 0.5	< 8
Br	6.4 ± 0.4	26 ± 1	7.5 ± 0.4	9.4 ± 0.4	13.5 ± 0.6	14.0 ± 0.8	9.0 ± 0.6	9.6 ± 0.6	10.2 ± 0.6	4.8 ± 0.4	4.3 ± 0.4
Ca	46 ± 8	86 ± 10	44 ± 15	22 ± 4	15 ± 5	94 ± 15	69 ± 8	12.5 ± 8.0	66 ± 20	14.8 ± 4.0	59 ± 7
Cl	450 ± 50	1390 ± 90	260 ± 25	36 ± 15	280 ± 35	2250 ± 30	320 ± 30	190 ± 30	111 ± 30	210 ± 22	3060 ± 30
Cu	< 1	2.4 ± 0.5	0.8 ± 0.1	< 0.5	< 2	1.1 ± 0.2	1.1 ± 0.2	4.0 ± 1.5	5.1 ± 1.9	1.1 ± 0.3	< 1
Dy	< 0.02	< 0.008	< 0.003	< 0.006	< 0.01	< 0.02	< 0.006	0.048 ± 0.007	< 0.007	< 0.007	< 0.005
I	0.90 ± 0.09	1.71 ± 0.15	1.23 ± 0.06	0.99±0.06	1.90 ± 0.09	1.47±0.30	2.9±0.1	2.7 ± 0.1	2.1 ± 0.1	1.52 ± 0.3	1.52 ± 0.1
In	< 0.005	< 0.005	< 0.002	< 0.001	0.0037 ± 0.002	<0.006	0.0069±0.0014	0.0067±0.0014	0.0109±0.0017	0.0025±0.0008	0.0039±0.0008
Mg	<50	180 ± 40	80 ± 20	40 ± 15	<50	225 ± 70	55 ±10	<20	<50	30 ± 10	380 ± 80
Mn	0.59 ± 0.04	3.2 ± 0.2	1.49 ± 0.07	0.51 ± 0.03	0.92 ± 0.05	1.04±0.10	5.4±0.2	3.4 ± 0.2	3.2 ± 0.2	1.27 ± 0.1	2.3 ± 0.1
Na	330 ± 10	1140 ± 30	210 ± 7	44 ± 3	310 ± 10	1530±40	610±15	320 ± 10	154 ± 5	210 ± 10	2160 ± 10
Sn	<4	1.5 ± 0.4	5.8 ± 0.4	4.6 ± 0.4	< 3	8.1±0.9	< 0.5	< 8	13.5 ± 1.0	5.1 ± 2.0	6.8 ± 0.5
Ti	1.5 ± 0.8	7.9 ± 0.8	1.9 ± 1.1	1.9 ± 0.8	< 2	NA	9.4 ± 2.0	3.9 ± 1.1	6.0 ± 2.0	< 3	< 2
V	0.32 ± 0.03	2.7 ± 0.1	1.21 ± 0.06	0.25 ± 0.02	0.78 ± 0.09	0.79±0.05	9.8 ± 0.3	8.5 ± 0.6	10.8 ± 0.5	2.2 ± 0.1	0.37 ± 0.04
SO_4	970	4670	2220	1110	3210	3330	11100	10000	790	570	650

*Due to errors in volume measurements, the concentrations for samples 1,2, and 5 have uncertainties of ± 100, 35 and 80 percent respectively. Uncertainties in table are analytical only.

APPENDIX B

THE UTILITY OF TRAJECTORY ANALYSIS IN THE ARCTIC

Abstract

The relative error involved in the determination of a wind vector on the 500 mb pressure surface was determined. Analysed maps from the United States, Japan, U.S.S.R. and the Federal Republic of Germany were used as different sources of interpolated data. All source maps were drawn from the same data base. Wind vectors were derived at 8 points along the seventy and eighty degree north latitude circles from contour spacing and orientation. Results show that within this data sparse region each wind vector interpolated from observed wind data has as a single standard deviation about the mean $\pm$ 34% for wind speed and $\pm$ 27 degrees in azimuth for wind direction from the four data sources. Isobaric or mean layer trajectories constructed in this region all rely upon data interpolation from observations. Trajectories greater than a few days in length must be interpreted with caution in data sparse regions such as the Arctic.

INTRODUCTION

Since the first chemical evidence that the aerosol comprising Arctic haze was primarily anthropogenic in origin (Rahn and Heidam, 1981), the primary unresolved question has been what is the aerosol source region. The logical steps taken by most investigators has been to derive backward in time, trajectory analyses from the point of aerosol collection to determine the source of the air mass associated with the aerosol (Halter and Peterson, 1981; Miller, 1981; Barrie et al., 1981; Borys and Rahn, 1981). These trajectories have primarily been constructed on a constant pressure surface or were representative of a "mean layer", constrained either between two pressure surfaces or interpolated between them. In all cases, the same basic upper air wind observations are used. In the Arctic region, the data base becomes very sparse as a result of virtually no reporting stations over the Arctic

Ocean. Even if the question of whether these types of trajectories represent air motion in the region is ignored, the lack of data should cast some doubt on their accuracy.

APPROACH

This study was undertaken to determine what kind of uncertainty should be placed upon each step made in the construction of a trajectory that relies upon the interpolated wind fields in the Arctic. Since all trajectory schemes are dependent upon an interpolation scheme of one kind or another, analysed weather maps from four different countries were used as sources of wind data that had been interpreted from the same world wide observational network. The sources of the maps included the respective meteorological services of the United States, Japan, Soviet Union, and the Federal Republic of Germany. The maps from the United States were computer generated. The other maps were all hand drawn from plotted wind observations. Because of the difficulty in locating maps from all sources for the same period and pressure level, the study was restricted to an eleven day period in January of 1974 at the 500 mb level.

The most recent aerosol data from the Arctic suggest the primary transport of aerosol to the Arctic is in the lower troposphere. A study of trajectory utility at the 500 mb level may not appear appropriate. However, the data from this level should be of good quality since it is a mandatory level and all countries that produce upper air analyses include this level. In addition, because the planetary waves dominate the flow at this level, the day to day coherancy of the data should be good because of the relatively long periodicity of these waves. Thus any differances found among the different analyses should primarily be

due to small errors in analysis and not due to effects of atmospheric variability. As one moves down in the atmosphere, the flow becomes more dependent upon synoptic scale disturbances such as migrating cyclones. This would induce a much greater source of variability in the data and possibly create more error in the map analysis. Any errors found among the different 500 mb map analyses should be representative of the best case in the troposphere of minimal analysis error. The results presented should be looked upon as a minimum in the estimate of the errors involved in trajectory analysis.

METHOD

Figure 1 shows the location of the eight grid points from which wind vector data were collected from each of the four data sources for the period 1-11 January 1974 on maps valid at 0Z. This provided more than eighty data points for each grid location from each data source. This resulted in more than 320 wind vector analyses from which the results were derived. At each grid point, wind direction was determined from the contour gradient and the orientation of a line tangential to the gradient with respect to true north. The winds at this level are assumed to be geostrophic and parallel to the contours. Wind speed should therefore be inversely proportional to the contour spacing for a given contour interval. All the wind speed data were collected as a ΔN representative of the distance in kilometers between contours normalized to an interval of 60 meters.

All the data are summarized in Table 1. The four values at each grid point for ΔN and azimuth were averaged and a standard deviation determined using standard statistical methods. It is realized that these are small populations from which to derive a standard deviation.

TABLE 1

A compilation of wind speed and direction data from eight arctic positions for the period 1-11 January, 1974.

GRID POINT:		1		2		3	
DATE	SOURCE	WIND SPEED AN (km)	WIND DIRECTION (deg. az.)	WIND SPEED AN (km)	WIND DIRECTION (deg. az.)	WIND SPEED AN (km)	WIND DIRECTION (deg. az.)
1 Jan 74	USSR	226	020	1521	180	1004	247
	UK	333	031	951	160	539	253
	GER	403	000	1111	191	463	250
	USA	310	357	1408	163	1150	265
	MEAN (1σ)	318(73)	012(16)	1248(263)	174(15)	789(339)	249(16)
2 Jan 74	USSR	281	007	445	172	515	281
	UK	155	000	590	515	444	255
	GER	222	011	699	164	361	259
	USA	236	010	523	130	345	259
	MEAN (1σ)	224(52)	007(005)	564(108)	167(30)	16(79)	264(12)
3 Jan 74	USSR	339	322	562	055	328	238
	UK	320	323	539	135	317	240
	GER	292	327	833	149	667	225
	USA	253	329	419	118	805	253
	MEAN (1σ)	301(37)	325(3)	588(175)	114(41)	129(245)	239(11)
4 Jan 74	USSR	246	284	(1404)	(190)	936	321
	UK	216	265	380	034	1388	152
	GER	130	285	calm	---	1162	156
	USA	316	285	790	340	949	005
	MEAN (1σ)	277(77)	280(10)	858(515)	191(163)	11.0(212)	181(97)
5 Jan 74	USSR	129	331	281	307	725	152
	UK	127	319	377	311	292	337
	GER	139	340	1185	316	292	180
	USA	---	---	---	---	---	---
	MEAN (1σ)	132(6)	330(11)	614(497)	311(5)	4 6(250)	223(100)
6 Jan 74	USSR	234	355	866	272	351	167
	UK	130	359	539	289	447	142
	GER	148	001	394	270	426	141
	USA	158	359	477	279	575	184
	MEAN (1σ)	168(46)	359(3)	569(207)	278(9)	450(93)	159(21)
7 Jan 74	USSR	328	359	474	212	878	122
	UK	187	013	380	200	488	164
	GER	259	359	500	232	695	147
	USA	184	001	489	211	747	133
	MEAN (1σ)	240(68)	273(173)	461(55)	214(13)	702(162)	142(18)
8 Jan 74	USSR	480	356	357	180	585	211
	UK	228	337	514	188	450	201
	GER	199	305	417	207	353	203
	USA	201	341	293	182	351	195
	MEAN (1σ)	277(136)	335(21)	395(94)	189(13)	511(135)	203(7)
9 Jan 74	USSR	298	306	357	309	1 58	180
	UK	187	313	292	304	09	168
	GER	194	303	407	291	53	201
	USA	241	302	339	299	26	170
	MEAN (1σ)	230(51)	306(5)	349(48)	301(8)	76 (265)	180(15)
10 Jan 74	USSR	316	281	456	221	1 90	058
	UK	409	268	184	202	935	165
	GER	509	299	213	207	512	113
	USA	402	285	235	205	511	119
	MEAN (1σ)	409(79)	283(13)	272(124)	209(8)	91: (628)	114(44)
11 Jan 74	USSR	878	292	205	205	[illegible]	---
	UK	491	301	159	199	476	181
	GER	421	355	148	207	856	130
	USA	---	---	---	---	---	---
	MEAN (1σ)	597(246)	316(34)	171(30)	204(4)	66 (269)	156(36)

TABLE 1 cont.

GRID POINT:		4		5		6	
DATE	SOURCE	WIND SPEED ΔN (km)	WIND DIRECTION (deg. az.)	WIND SPEED ΔN (km)	WIND DIRECTION (deg. az.)	WIND SPEED ΔN (km)	WIND DIRECTION (deg. az.)
1 Jan 74	USSR	142	291	1189	203	273	063
	UK	158	256	1553	219	380	081
	GER	139	250	787	237	282	046
	USA	127	253	1309	200	416	060
	MEAN (1σ)	142(13)	263(19)	1210(320)	215(17)	338(71)	63(14)
2 Jan 74	USSR	129	300	421	224	761	063
	UK	114	306	726	239	723	215
	GER	652	172	295	232	1005	018
	USA	132	301	1642	239	970	201
	MEAN (1σ)	257(264)	270(65)	771(608)	234(7)	865(143)	124(99)
3 Jan 74	USSR	105	302	830	150	142	305
	UK	105	301	602	155	957	227
	GER	134	299	386	260	*calm*	---
	USA	109	297	1523	189	1094	090
	MEAN (1σ)	113(14)	300(2)	493(291)	150(109)	731(515)	207(109)
4 Jan 74	USSR	105	321	711	025	225	129
	UK	228	313	1157	031	558	135
	GER	144	313	1023	057	381	116
	USA	184	307	595	320	476	136
	MEAN (1σ)	165(53)	314(6)	872(262)	019(42)	410(143)	129(9)
5 Jan 74	USSR	246	305	391	273	344	049
	UK	184	293	338	276	352	055
	GER	232	301	286	295	381	098
	USA	---	---	---	---	---	---
	MEAN (1σ)	221(33)	300(6)	338(53)	281(12)	359(19)	067(27)
6 Jan 74	USSR	328	283	356	243	711	024
	UK	418	295	225	236	682	351
	GER	194	262	262	242	762	004
	USA	172	279	217	249	595	004
	MEAN (1σ)	278(116)	280(14)	265(64)	243(5)	688(70)	006(014)
7 Jan 74	USSR	152	318	201	208	261	024
	UK	174	310	266	200	333	028
	GER	160	309	233	213	314	044
	USA	118	217	286	208	309	035
	MEAN (1σ)	151(24)	289(48)	247(37)	207(005)	304(31)	033(009)
8 Jan 74	USSR	176	332	344	213	403	018
	UK	159	324	323	218	263	360
	GER	176	365	386	215	290	026
	USA	126	322	315	194	345	032
	MEAN (1σ)	159(24)	336(20)	342(32)	210(11)	325(62)	019(014)
9 Jan 74	USSR	222	360	261	235	261	025
	UK	238	335	460	242	447	008
	GER	162	351	390	214	295	029
	USA	374	346	327	239	208	013
	MEAN (1σ)	249(90)	348(10)	360(85)	233(013)	303(103)	019(10)
10 Jan 74	USSR	456	315	320	235	379	001
	UK	285	335	317	215	514	003
	GER	241	328	333	215	571	012
	USA	290	345	303	240	446	352
	MEAN (1σ)	318(95)	331(13)	318(12)	226(13)	478(83)	002(8)
11 Jan 74	USSR	257	296	284	215	142	053
	UK	323	305	269	191	187	006
	GER	486	301	233	199	238	028
	USA	---	---	---	---	---	---
	MEAN (1σ)	355(118)	301(5)	262(26)	202(12)	189(48)	29(24)

TABLE 1 cont.

GRID POINT:		7		8	
DATE	SOURCE	WIND SPEED AN (km)	WIND DIRECTION (deg. az.)	WIND SPEED AN (km)	WIND DIRECTION (deg. az.)
1 Jan 74	USSR	439	157	356	323
	UK	253	090	2171	326
	GER	366	151	1758	055
	USA	708	091	1785	343
	MEAN (1σ)	442(193)	122(37)	1518(797)	2[illegible](138)
2 Jan 74	USSR	456	153	573	354
	UK	1046	114	1073	355
	GER	242	271	757	007
	USA	476	180	1041	002
	MEAN (1σ)	555(344)	180(67)	861(238)	[illegible]9(6)
3 Jan 74	USSR	178	240	830	329
	UK	355	255	913	289
	GER	276	229	966	330
	USA	333	235	1261	323
	MEAN (1σ)	286(79)	240(11)	993(188)	3[illegible]5(18)
4 Jan 74	USSR	284	247	853	023
	UK	317	233	1056	215
	GER	295	219	624	003
	USA	268	230	654	360
	MEAN (1σ)	291(21)	232(12)	791(196)	3[illegible]5(74)
5 Jan 74	USSR	474	207	486	030
	UK	266	217	406	028
	GER	224	200	376	036
	USA	---	---	---	---
	MEAN (1σ)	321(134)	208(9)	423(57)	0[illegible]5(008)
6 Jan 74	USSR	782	166	480	041
	UK	418	204	235	028
	GER	381	180	1666	005
	USA	354	163	303	023
	MEAN (1σ)	484(201)	178(19)	671(671)	02[illegible](015)
7 Jan 74	USSR	474	253	249	123
	UK	257	242	197	078
	GER	721	243	362	102
	USA	387	246	292	080
	MEAN (1σ)	460(196)	246(005)	275(70)	0[illegible]6(021)
8 Jan 74	USSR	474	305	273	078
	UK	228	276	222	075
	GER	405	257	219	081
	USA	345	307	208	093
	MEAN (1σ)	363(104)	286(024)	231(29)	08[illegible]2(008)
9 Jan 74	USSR	225	247	344	078
	UK	184	275	219	063
	GER	405	257	228	082
	USA	327	275	208	077
	MEAN (1σ)	285(100)	264(14)	250(63)	[illegible]5(8)
10 Jan 74	USSR	356	238	225	076
	UK	295	263	241	082
	GER	285	260	295	094
	USA	452	243	205	090
	MEAN (1σ)	347(77)	251(12)	242(39)	[illegible](8)
11 Jan 74	USSR	356	299	284	030
	UK	197	286	174	090
	GER	262	289	314	087
	USA	---	---	---	---
	MEAN (1σ)	272(80)	291(7)	257(74)	6[illegible](34)

These data are used here only as a first approximation of the spread of values of the wind speed and direction at a grid point. The mean and standard deviation are listed below each set of figures.

Table 2 is a summary of the wind speed and direction error by grid point from the four data sources. The values reported are one standard deviation of the derived wind speed and direction for each grid point using the standard deviations of the means calculated for each of the eleven days from Table 1. This is presented as percent of the mean for the wind speed and in degreees for the azimuth. At the bottom of the table is the average standard deviation using the eight values presented above for all grid points. This is a value that is applicable to the area covered by the grid point locations which in this case is considered to include all the area north of the 70 degree latitude circle. It is apparent that the error involved in the determination of a wind vector in the Arctic is fairly consistent over the entire region. The range of uncertainty in wind speed is 11% and of wind direction is 14 degrees. These values should be representative of an arctic- basin-wide uncertainty that should be applied to a trajectory for each time step constructed. The result of the application of such an uncertainty to a trajectory analysis will be demonstrated below.

Maps of the 500 mb pressure surface valid for 5-6 January 1980 were chosen as typical of conditions during mid-winter for the Arctic. An example trajectory was constructed backward in time beginning on the island of Spitsbergen located at approximately 79°N, 15°E. The method of constructing the trajectory is that same as that described by Saucier (1955). Wind speed and direction were determined directly from the map using contour spacing and orientation and assuming geostrophic flow.

TABLE 2

Arctic 500 mb wind speed and direction error Summary

Grid Point	OVERALL MEAN ONE STANDARD DEVIATION	
	WIND SPEED (%)	WIND DIRECTION (degrees)
1	28	27
2	35	28
3	37	34
4	35	20
5	33	21
6	26	31
7	37	20
8	37	31

AVERAGE STANDARD DEVIATION
FOR ALL GRID POINTS

WIND SPEED	WIND DIRECTION
± 34%	± 27 degrees azimuth

Maps valid at 12Z, 5 January and 0Z and 12Z, 6 January were used. Time steps of 6 hours were used to construct the trajectory thus allowing the contour configuration of two consecutive maps to input on a single 12 hour time step.

The first time step using both the speed and azimuth uncertainty produced an area of uncertainty encompassing the region from which an air parcel could have originated 6 hours earlier. This produced an area in the shape of a trapezoid. Subsequent steps had to use a less rigorous method. Since the following steps were dealing with an area with essentially an infinite number of points which had to be advanced a six hour time step, a carefully selected set of points were chosen from which to continue the analysis which would produce a good approximation of the true boundary of the area of uncertainty of the given time step. After one or two time steps, it became obvious that very few steps would have to be made before the outcome was clear.

RESULTS

The results of four, 6 hour backward time steps from Spitsbergen are depicted in Figure 2. Included in the figure is a simple linear trajectory constructed in 6 hour time steps for 24 hours as indicated by the line connecting heavy dots. Each dot represents one 6 hour time step. The outlined and hatched areas indicate the boundaries of uncertainty for each of the 6 hour time steps. The 500 mb maps valid for this period show closed low pressure centers located to the west of Greenland and the the northeast of Spitsbergen. The effect of the synoptic situation can be seen quite clearly in the way in which the trajectory uncertainty boundary evolves. Southerly flow around both low centers dominates the trajectories. This combination tends to elongate

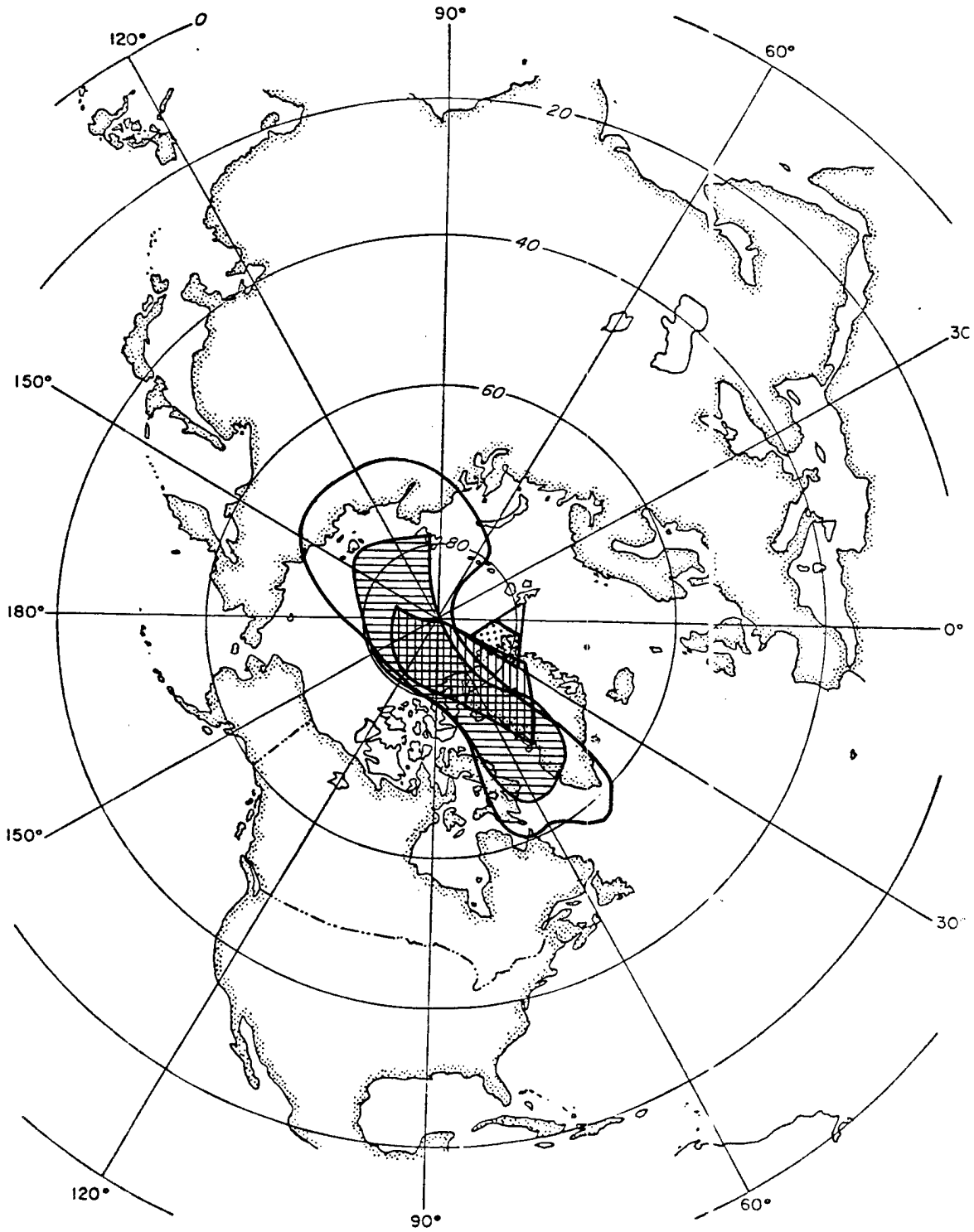


Figure 2: Results of four, 6 hour backward time steps beginning at the island of Spitzbergen at 12Z 6 January 1930 on the 500 mb surface. Heavy line and dots represents the simple trajectory. The areas of uncertainty for each time step are outlined and cross hatched.

the boundary of uncertainty until after only a 24 hour period the north-south extent is over 6000 km. It is emphasized that based upon the potential error in determining a wind vector, the air parcel reaching Spitsbergen at 12Z 6 January 1980 could have originated anywhere within the outer boundary depicted in figure 2. This analysis does not imply anything about atmospheric processes such as diffusion, convergence or divergence.

CONCLUSIONS

The purpose of this study was to ascertain the confidence with which isobaric trajectories can be constructed and interpreted in data sparse regions such as the Arctic. Certainly this study has some limitation in its interpretation because it uses the very technique which is under scrutiny. The trajectory presented as an example only represents a single case study. Certainly other cases will produce results with varying degrees of error involved in their construction. As a first approximation it is apparent that caution should be exercised when interpreting trajectory analyses, especially those that extend through many time steps over many days. An appropriate use of trajectory analyses would be in conjunction with other atmospheric measurements of a conservative or semi-conservative property in the same area of interest including physical, thermodynamic and/or chemical parameters. Isobaric trajectories have been used extensively in the past, and new methods are being devised to construct similar types, such as mean layer trajectories, by computer. Caution is advised in attempts to further refine these kinds of analyses when the ultimate limitation is the quality of the input data.

Three conclusions were drawn from this study: 1) Uncertainties in the wind data will produce uncertainties in the final trajectories that exceed the magnitude of the trajectory itself; 2) Trajectories greater than 3-5 days in length, especially in the lower troposphere, should be interpreted with extreme caution; 3) There is no substitution for manual trajectory construction unless the goal is to produce a climatologically significant set of data.

REFERENCES

Barrie, L. A., R. M. Hoff and S. M. Daggupaty (1981) The influence of mid-latitudinal pollution sources on haze in the Canadian arctic. Atmospheric Environment, 15, 1407-1419.

Borys, R. D. and K. A. Rahn (1981) Long-range atmospheric transport of cloud-active aerosol to Iceland. Atmospheric Environment, 15, 1491-1501.

Halter, B. C. and J. T. Peterson (1981) On the variability of atmospheric carbon dioxide concentration at Barrow, Alaska, during summer. Atmospheric Environment, 15, 1391-1399.

Miller, J. M. (1981) A five-year climatology of five day back trajectories from Barrow, Alaska. Atmospheric Environment, 15, 1401-1405.

Rahn, K. A. and N. Z. Heidam (1981) Progress in Arctic air chemistry, 1977-1980: A comparison of the first and second symposia. Atmospheric Environment, 15, 1345-1348.

Saucier, W. J. (1955) Principles of Meteorological Analysis, University of Chicago Press.

Acknowledgements

Supported by the Office of Naval Research (Contract N00014-76-C-0435) and by the National Science Foundation (Grant No. ATM 78-19261).

APPENDIX C

Atmospheric Environment, Vol. 15, No. 8, pp. 1491–1501, 1981
Printed in Great Britain.

0004-6981/81/081491-11 $02.00/0

LONG-RANGE ATMOSPHERIC TRANSPORT OF CLOUD-ACTIVE AEROSOL TO ICELAND*

RANDOLPH D. BORYS
Department of Atmospheric Science, Colorado State University, Ft. Collins, Colorado 80523, U.S.A.

and

KENNETH A. RAHN
Graduate School of Oceanography, University of Rhode Island, Kingston, Rhode Island 02881, U.S.A.

(*First received* 7 *October* 1980 *and in final form* 9 *January* 1981)

Abstract—During a study of cloud-condensation nuclei (CCN), condensation nuclei (CN) and various chemical constituents of the aerosol at Irafoss, Iceland, an episode of long-range transport of CCN and pollution aerosol from Eurasia was observed. This air was enriched in CCN by factors of 2–5 and in pollution aerosol by factors of 5–10 compared to near-background air from the west Atlantic and Arctic. CCN spectra of the aged European air were enhanced primarily in the largest particles, those active at the lowest supersaturations. The CCN appeared to be explained largely by nonmarine $SO_4^=$ and some yet-unidentified constituent of the aerosol, possibly organic matter. From these results, one may predict that anthropogenic CCN can be transported throughout the Arctic in a fashion similar to that already observed for $SO_4^=$.

INTRODUCTION

Recent studies of aerosol and precipitation in and aroupd the Arctic have revealed trace-element concentrations considerably greater than those of typical remote regions, particularly during the winter half-year. In most of these cases, the high concentrations are attributable to long-range transport of anthropogenic aerosol from midlatitudes (Rahn *et al.*, 1977; Gjessing, 1977; Rahn and McCaffrey, 1979a, 1979b, 1980; Rahn, 1980; Larssen and Hanssen, 1980; Ottar, 1980; Barrie *et al.*, 1981). The main source regions and meteorological aspects of transport to the Arctic have been dealt with by Rahn and McCaffrey, 1979b, 1980; Rahn, 1980, 1981a, 1981b). Eurasia seems to be the principal source region; transport to the Arctic appears to be primarily between the Icelandic low and the Asiatic high, at low elevation (well below 5 km), and of duration 10–20 days.

Anthropogenic aerosol in the Arctic atmosphere may have radiative, depositional and nucleational effects. Direct radiative effects have been dealt with elsewhere (Shaw and Stamnes, 1980); the nature and amounts of deposition of aerosol in the Arctic are also under study (Weiss *et al.*, 1978; Rahn and McCaffrey, 1979a, Rahn, 1980; Rahn, 1981c). One of the most efficient ways for an aerosol to affect the radiation balance of an area, hence affect its climate, is to nucleate droplets or ice crystals in the atmosphere. These so-called "cloud-active aerosols" are of two types, cloud-condensation nuclei (CCN) and ice nuclei (IN). The CCN are hygroscopic particles which form hazes at high relative humidities or clouds and fogs at low supersaturations; the IN may have quite different properties and are less understood at present. Changes in the concentrations of CCN and IN have been related to changes in the microstructure of clouds in an air mass (Twomey, 1959; Fitzgerald and Spyers-Durbin, 1973; Hobbs *et al.*, 1980). Such changes would be expected to change the size distributions of cloud droplets as well as the relative numbers of ice crystals in the cloud, which in turn should change the radiative properties of the cloud. Inasmuch as one of the major components in the radiation balance of the Arctic is persistent stratus cloud (Fletcher, 1965), any modification of the radiative properties of Arctic clouds by anthropogenic aerosol may have palpable climatic consequences.

Very little is known about cloud active aerosol in the Arctic. Radke *et al.* (1976) measured CCN and IN at Barrow, Alaska during March 1970 and found, to their surprise, that much of the time the concentrations of CCN were 100–500 cm^{-3} at 1% SS, several times higher than values they had measured on remote Mt. Olympus, Washington. Peak values of CCN (2000 cm^{-3}) were typical of urban areas. There were no obvious local sources for these CCN. Jayaweera and Ohtake (1973) reported IN concentrations at Barrow for September 1971 and April 1972 as a function of temperature. Other than these reports, we are not aware of any CCN or IN data for the Arctic. In an effort to remedy this, we have begun a systematic, large-scale study of CCN and IN at selected sites in and around the Arctic, in a way designed to determine the importance of long-range transport from midlatitudes. Summer and winter experiments have been carried out

* Paper presented at the Second Symposium on Arctic Air Chemistry, University of Rhode Island, Kingston, RI 02881, U.S.A., 6–8 May 1980.

at six sites: Barrow, Alaska; Pangnirtung, NWT; and Ny Ålesund, Spitsbergen (Arctic); Poker Flat, Alaska and Irafoss, Iceland (sub-Arctic); and Narragansett, Rhode Island (midlatitude source region). Locations of these sites are shown in Fig. 1.

Iceland was chosen to represent near-Arctic aerosol along North Atlantic pathways from North America and Europe, rather than true Arctic aerosol, which is presently being studied at more northerly sites such as Barrow, Alaska (Rahn and McCaffrey, 1980), Mould Bay, NWT (Barrie *et al.*, 1981) and Bear Island and Spitsbergen (Larssen and Hanssen, 1980). This article reports results from the summer study in Iceland, which illustrate the approach taken at the other sites, and serve as a case study of long-range transport. Data from other sites and other seasons will be reported as they become available.

One of the important features of this study was the concomitant measurements of trace constituents of the aerosol, such as $SO_4^=$, V, Mn, Al, etc., whose long-range transport in and around the Arctic has now been studied considerably. By relating the behavior of the cloud-active aerosol to these other constituents, among which are both primary and secondary as well as natural and anthropogenic species, it should be possible to elucidate the origin, transport, and perhaps also the chemical identity of the cloud-active aerosol. This is, of course, an ambitious goal, for aerosols and gases form a highly dynamic system, especially when derived from polluted continental regions. There are certainly limits to what can be deduced about the evolution of an aerosol during several days' transport from measurements at a single surface site. We feel, however, that coupling chemical and physical techniques can provide a surprisingly great amount of information on the nature and importance of such transport.

There is potential ambiguity in the term "long-range transport" when applied to secondary substances such as $SO_4^=$. When air comes from a well-defined source area, such as a strongly polluted continent, primary pollutants like V or Mn detected at the receptor site have been transported the entire distance, whereas secondary pollutants have been partially formed *en route*, hence have travelled shorter distances. In this article, "long-range transport" is interpreted broadly,

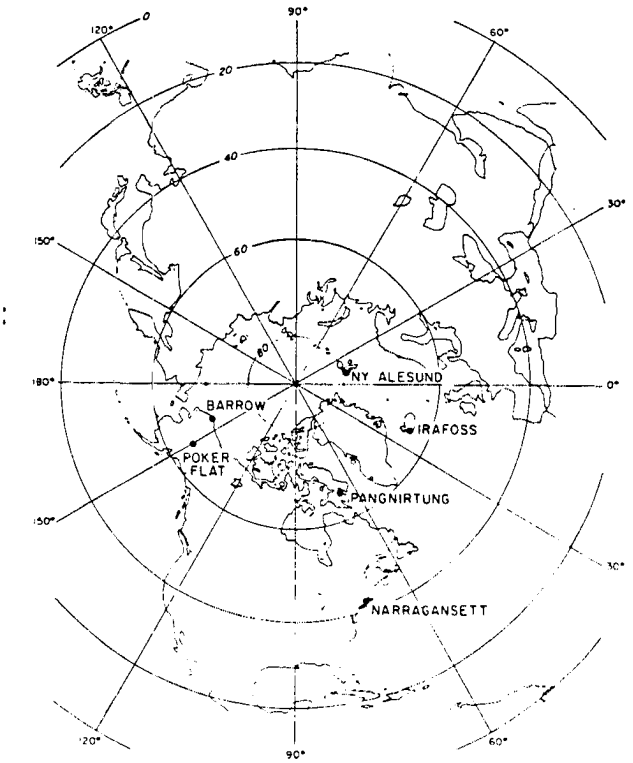


Fig. 1. Sampling sites for study of cloud-active aerosol in the Arctic.

to include secondary pollutants, provided that their precursor gases had the same origin as did the primary aerosol pollutants.

SAMPLING AND ANALYSIS

The sampling site was Irafoss hydroelectric station, about 30 km east of Reykjavik, Iceland. Local sources of aerosol were minimal, and were limited to sparse traffic and small deserts to the north. Iceland as a whole has nearly no heavy industry to produce aerosol; the only significant fossil-fuel combustion is by automobiles, because virtually all domestic and industrial energy comes from hydroelectric and geothermal sources. Thus, Iceland is an excellent location from which to study long-range transport of aerosol over the North Atlantic.

CCN were activated with a Mee Industries Model 130 cloud-condensation nucleus counter, and were photographed with a 5 mW He–Ne laser and 35 mm camera. CCN concentrations were determined visually from the negatives. Spectra for supersaturations of 0.20, 0.50, 1.0 and 2.0% were determined each 12 h. Details of this instrument and its performance have been reported by Borys (1980). IN concentrations were determined from particles collected on 0.2 μm Nuclepore filters, using a modified static diffusion chamber and a technique similar to that described by Stevenson (1968). In addition, condensation nuclei (CN) were measured with a Gardner Small Particle Counter several times a day.

Aerosol for chemical analysis was collected on continuous 24 h high-volume filters. A series of elements (Na, Mg, Cl, Br, I, Al, Ti, Mn, V, etc.) was determined by nondestructive neutron activation; sulfate was determined by a modified version of the turbidimetric technique of Tabatabai (1974). In addition, 0.2 μm Nuclepore filters were taken to determine particle-size distributions by scanning electron microscopy and elemental composition of single particles by energy-dispersive X-ray spectrophotometry.

In this article, CCN, CN and trace-element results will be discussed. Results of the other analyses will be reported later.

RESULTS

Figure 2 gives 24 h means of some of the data obtained for 17–26 August 1979 at Irafoss. CCN concentrations are given for the intermediate supersaturation of 0.2%; counts at other supersaturations were generally parallel. Three indicators of natural aerosol are given—marine Na for sea salt, Al for soil dust, and I for secondary marine aerosol. Marine, or

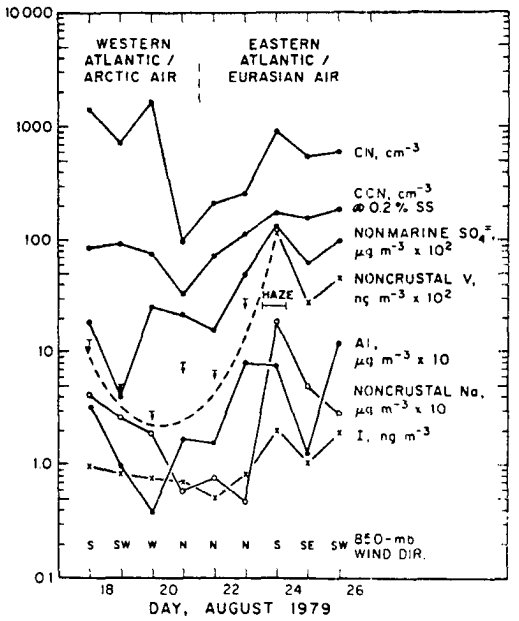


Fig. 2. Atmospheric concentrations of CN, CCN, and several chemical constituents of the aerosol at Irafoss, Iceland, August 1979. All data are 24 h means.

noncrusta , Na was derived from total Na and Al by the following equation

$$Na_{noncrustal} = Na_{total} - \left(\frac{Na}{Al}\right)_{crust} Al_{total} \quad (1)$$

where Al and noncrustal Na are presumed to be derived exclusively from the crust and the sea, respectively. The Na/Al ratio of bulk Icelandic rock, 0.2, was used (C. K. Unni, personal communication). (Selection of the proper Na/Al ratio for use in Equation (1) was critical for samples 4–6, which had low Na and high Al. A value of 0.25, for example, was too high, because it gave unrealistically low concentrations of marine Na. Thus, the Na/Al ratio of mean crustal rock, 0.35 (Mason, 1966) could not be used here.) The sea salt obviously came from the North Atlantic, but without size-distribution data for the salt particles it is impossible to say from how far away they originated. The soil dust may have had a local or a distant origin; V/Al ratios which supported a local origin are presented below. The significance of the I data is also discussed below.

Two indicators of pollution-derived aerosol, nonmarine $SO_4^=$ and noncrustal V, are also shown in Fig. 2. Nonmarine $SO_4^=$ was calculated from total $SO_4^=$ by the following equation:

$$SO_{4\ nonmarine}^{=} = SO_{4\ total}^{=} - \left(\frac{SO_4^=}{Na}\right)_{sea\ salt} (Na)_{noncrustal} \quad (2)$$

The $SO_4^=/Na$ ratio of sea-salt aerosol used here was 0.25, the value for bulk seawater (Pytkowicz and Kester, 1971). Laboratory generation of sea-salt aerosol from seawater (Gravenhorst, 1978) has yielded values between 0.22 and 0.28 for this ratio, confirming that there is no fractionation of $SO_4^=$ relative to Na at the source. Nonmarine $SO_4^=$ was 37–98% of the total $SO_4^=$.

Nonmarine $SO_4^=$ is associated with aerosol other than that produced by breaking bubbles at the sea surface. It is submicron and presumably secondary (Gravenhorst, 1978). Photochemical oxidation of precursors such as COS, CS_2, $(CH_3)_2S$ and H_2S, a portion of which may be marine-derived, can account for approximately 0.1 $\mu g\ m^{-3}$ nonmarine $SO_4^=$ (Sze and Ko, 1980). These concentrations are typically found over remote oceans and in other background regions of the atmosphere. Over less remote oceans, however, such as the midlatitude North Atlantic, concentrations of nonmarine $SO_4^=$ can be several times higher than this (Gravenhorst, 1978), in which case the $SO_4^=$ probably comes from oxidation of pollution-derived SO_2.

Noncrustal V in atmospheric aerosol has a single major (pollution) source, combustion of residual oil (Zoller *et al.*, 1973), and has been used extensively as an indicator of transport of aerosol from midlatitudes to the Arctic (Rahn and McCaffrey, 1979b, 1980; Rahn, 1980). The crustal component of V is subtracted from the total V in a parallel fashion to noncrustal Na:

$$V_{noncrustal} = V_{total} - \left(\frac{V}{Al}\right)_{crust} Al_{total} \quad (3)$$

V/Al ratios for global average crustal rock are normally used for this type of subtraction, but in these samples the V/Al ratio never fell below about double the bulk crustal ratio, even when the concentrations of V and Al were high enough that we were sure they were both derived from local soils (see Table 1). This is one of the first problems we have encountered with global crustal ratios applied to remote-area aerosols. The consistency of the V/Al ratio in samples 1–6 implied that local soils were influencing even these early samples and that the V/Al ratio of the Icelandic soils was more than double that of global crust. This was perhaps reasonable, because Icelandic rock is basaltic and V is generally enriched in basalts by a factor of two compared to mean crustal rock (Gmelin, 1968). For the V/Al ratio in Equation (3), we used the mean from samples 1–6, $(3.6 \pm 0.21) \times 10^{-3}$; this ratio is close to that of bulk Icelandic rock, 4.25×10^{-3} (C. K. Unni, personal communication).

Noncrustal Mn (hereafter referred to simply as Mn) is often a good indicator of pollution aerosol; its values were calculated as well (see Table 2), but it was not plotted in Fig. 2 because only sample 7 had a detectable value. The local Mn/Al ratio also appeared to be significantly different from the global value; a value of $(22.8 \pm 2.5) \times 10^{-3}$ was derived from samples 1–6 and 8–9 and used in the subtraction. This value compared well with the 19.6×10^{-3} for Icelandic rock; it was roughly double the global crustal ratio.

Three representative CCN spectra are presented in Fig. 3, each the mean of three determinations. Error bars represent experimental spread of the data.

DISCUSSION

On the basis of data shown in Fig. 2 and the meteorological discussion below, the Iceland experiment could be divided into two parts, a near-background period during the first four days and an episode of transport of European aerosol during the last five days. The data of Fig. 2 were insufficient to establish this division, however. They showed several different patterns of variation: broad minima during the middle three days for CN and noncrustal Na (hereafter called simply Na), a highly irregular pattern for Al, somewhat higher concentrations during the second half for CCN and IN, and markedly higher concentrations during the second half for nonmarine $SO_4^=$ and noncrustal V (hereafter referred to simply as $SO_4^=$ and V). Both CN and CCN had sharp minima on the fourth day, but divergent patterns otherwise: the CN peaked during the first three days whereas the CCN peaked during the last three days. Occasional one-day minima were seen for other constituents such as $SO_4^=$ and Al.

The local 850 mb wind direction helped to interpret several features of this plot having to do with the

Table 1. Concentrations (ng m^{-3}) and ratios of Al, Mn and V in the Icelandic aerosol of August 1979. All uncertainties 1σ

Sample	Dates (Aug. 1979)	Al	Mn	Mn/Al ($\times 10^{-3}$)	V	V/Al ($\times 10^{-3}$)	Noncrustal Mn	Noncrustal V	Noncrustal Mn / Noncrustal V
1	17–18	346 ± 20	8.37 ± 0.60	24.2 ± 2.2	1.22 ± 0.09	3.53 ± 0.33	< 0.9	< 0.13	
2	18–19	97 ± 8	2.68 ± 0.20	27.6 ± 3.1	0.338 ± 0.030	3.48 ± 0.42	0.47 ± 0.28	< 0.05	
3	19–20	38 ± 5	0.81 ± 0.15	21.2 ± 4.8	0.148 ± 0.020	3.89 ± 0.73	< 0.2	< 0.03	
4	20–21	168 ± 10	3.96 ± 0.30	23.6 ± 2.3	0.642 ± 0.060	3.82 ± 0.42	< 0.5	< 0.08	
5	21–22	155 ± 10	3.09 ± 0.20	19.9 ± 1.8	0.524 ± 0.050	3.38 ± 0.39	< 0.4	< 0.07	
6	22–23	807 ± 50	16.7 ± 1.0	20.7 ± 1.8	3.02 ± 0.20	3.74 ± 0.34	< 1.7	< 0.30	
7	23–24	752 ± 40	22.2 ± 1.4	29.5 ± 2.4	3.95 ± 0.30	5.25 ± 0.49	5.05 ± 1.80	1.21 ± 0.35	4.2 ± 1.9
8	24–25	126 ± 9	3.02 ± 0.20	24.0 ± 2.3	0.744 ± 0.060	5.90 ± 0.64	< 0.4	0.28 ± 0.07	< 1.5
9	25–26	1196 ± 60	26.6 ± 1.5	22.2 ± 1.7	4.83 ± 0.30	4.04 ± 0.32	< 2.3	0.48 ± 0.42	< 4.8
	Mean of 1–6, 8–9			22.8 ± 2.5	Mean of 1–6	3.64 ± 0.21			
	Mean Icelandic rock (C. K. Unni, personal communication)			19.6		4.25			
	Mean crustal rock (Mason, 1966)			11.7		1.66			
	Mean Soil (Vinogradov, 1959)			11.9		1.40			

Table 2. $SO_4^=/V$ ratios in Icelandic aerosol of August 1979

Sample	Dates, August 1979	Nonmarine $SO_4^=$, ng m^{-3}	Noncrustal V, ng m^{-3}	$SO_4^=/V$ ($\times 10^3$)
1	17–18	184 ± 33	< 0.13	> 1.42
2	18–19	40 ± 12	< 0.05	> 0.80
3	19–20	250 ± 30	< 0.03	> 8.33
4	20–21	212 ± 17	< 0.08	> 2.65
5	21–22	157 ± 13	< 0.07	> 2.24
6	22–23	500 ± 37	< 0.30	> 1.67
7	23–24	1343 ± 106	1.21 ± 0.35	1.11 ± 0.33
8	24–25	636 ± 51	0.28 ± 0.07	2.27 ± 0.60
9	25–26	999 ± 68	0.48 ± 0.42	2.08 ± 1.83

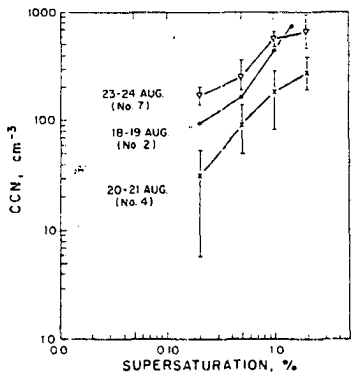


Fig. 3. Three typical CCN spectra from Irafoss, Iceland, August 1979. Each spectrum is the mean of three determinations during 24 hours. Error bars are based on experimental scatter.

shorter-lived constituents of the aerosol (the supermicron and Aitken particles), which apparently were quasi-local in origin. During the first three days, the winds were from the SW and the supermicron aerosol was relatively marine in character, as evidenced by a Na/Al ratio greater than unity. This was not surprising, as Irafoss is in SW Iceland, not far from the sea. During the second three days, when the local wind came from the north and had to pass over most of Iceland before reaching Irafoss, the supermicron aerosol was more continental, with the Na/Al ratio less than unity. This lower ratio was the result of two influences: low Na concentrations from dilution/removal of marine aerosol at the north coast of Iceland and higher Al concentrations from Iceland in general and from small, stony deserts 50–60 km NNW of Irafoss (dust clouds were occasionally observed in the direction of these deserts during the sampling). During the last three days, when local winds came from the SW-SE, the supermicron aerosol could be classified mixed marine-continental, because Na and Al had relatively high concentrations of variable dominance. The high Al of the last day was probably a purely local effect, from weekend traffic on dirt roads near the sampling site.

The influence of Reykjavik during the first three days, when it was generally upwind of the sampling site, was apparently seen in the high CN of that entire period and possibly in the high Al of the first day. The effect on CN was interesting, for it suggested that the CN count of remote areas may be a more sensitive indicator of local sources than of long-range transport. A similar effect has been reported recently for Barrow, Alaska (Peterson *et al.*, 1980), where transport from the north could be seen clearly in light scattering (a measure of the submicron aerosol) but not in CN count. This behavior of CN is probably a result of their short atmospheric lifetimes, caused by rapid coagulation of the Aitken nuclei which make up the vast majority of CN numbers.

The longer-lived submicron aerosol of Fig. 2 (CCN, $SO_4^=$ and V) required more extensive meteorological analysis and construction of air-mass trajectories of several days' duration to explain its variations properly. As discussed below, these results showed clearly that the submicron aerosol had quite different origins than did the supermicron and Aitken aerosol. During the first day of sampling, air at 850 mb (1.5 km) came to Iceland from the southwest, under the combined influence of a trough over southern Greenland and the Bermuda high, which extended quite far north at that time. By the second day of sampling, a trough was beginning to form over Iceland; it deepened and progressed eastward, so that by the fourth day of sampling the air-flow to Iceland came from the north. The trough remained stationary and deepened into a closed low during the fourth and fifth days of sampling, during which time it advected air around its backside from Europe towards Iceland in a manner very similar to that proposed for flow into the Arctic around the more northerly placed Icelandic low of winter (Rahn, 1980). By the last three days of sampling, the low had dissipated and high pressure, low-wind conditions dominated.

Trajectories for each of the nine days are shown in Fig. 4. They were calculated in 12 h time steps at 850 mb, using observed wind speeds and analysed contours for wind direction. They are all for five days, except for the first (4.5 d) and the fourth (3d). They clearly divided the sampling period into two parts, the

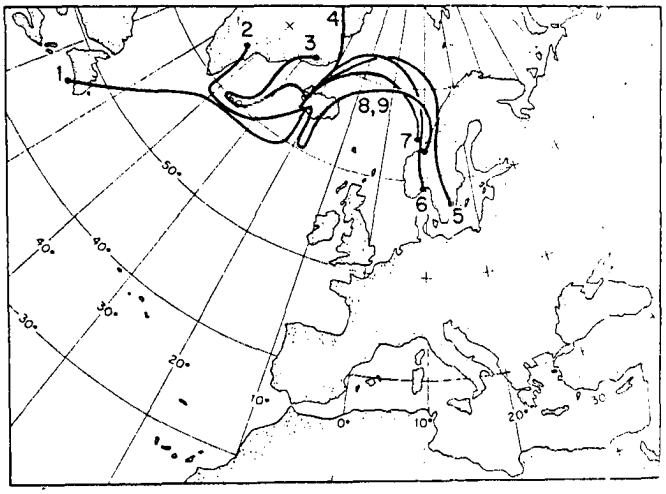


Fig. 4. 850 mb air-mass trajectories to Iceland for each of the days of this study. Numbers refer to the day of arrival at Irafoss. All trajectories are five days long, except for the first (4.5d) and the fourth (3d).

first four days when the air came from the west (called West Atlantic/Arctic air in Fig. 2) and the last five days when the air came from the east (called East Atlantic/Eurasian air in Fig. 2). During this first period, concentrations of the various constituents of the submicron aerosol were very low and roughly constant: $SO_4^=$ was at near-background levels of 0.04 to 0.3 $\mu g\,m^{-3}$, V was below its detection limit of 0.03 to 0.13 $ng\,m^{-3}$, and CCN concentrations were near the upper limit of those found in other remote regions and considered to be background (Jiusto, 1966; Blanchard, 1969; Twomey and Wojciechowski, 1969; Hobbs, 1971; Elliott and Egami, 1975; Radke *et al.*, 1976; Hoppel, 1979); interestingly, they are also roughly the concentrations of cloud droplets in Arctic summer stratus (Jayaweera and Ohtake, 1973).

Thus, all evidence pointed to a background or near-background character of the submicron aerosol of the first period. For samples 2–4, where the air came from the Arctic, this is perhaps not surprising. For sample 1, where the air came from the direction of North America, it may mean either that the air had a marine origin in the subtropical Bermuda high, or that it came from North America but was cleansed and diluted *en route*. At present we do not know which is the correct explanation. Transport from North America would have taken 5–7 days at 850 mb to cover the 5000–6000 km. While such distances for efficient transport of aerosol in midlatitudes are not impossible, even in summer, they may be infrequent over oceans (Rahn, 1980). In this case, the V and Mn generated from Icelandic soils effectively masked any small residues of transatlantic transport from North America; the concentrations of $SO_4^=$ were low enough so that they alone could not be used to discriminate between tropospheric and North American pollution sources. The Icelandic crustal aerosol could not mask the much stronger subsequent transport from Eurasia, however.

During the last five days, when the trajectories were strikingly similar to one another and from midlatitude Eurasia, concentrations of the submicron $SO_4^=$, V and CCN had their maxima, and CN concentrations had a relative maximum. The absolute maxima of $SO_4^=$ and V on the seventh day, 1.34 $\mu g\,m^{-3}$ and 1.21 $ng\,m^{-3}$, respectively, were high enough to strongly suggest pollution sources. They were also quite compatible with atmospheric transport on the scale of several thousand km (Rahn and McCaffrey, 1979b, 1980). On this day, haze was also observed at Irafoss. Thus, the submicron aerosol of this second period showed strong evidence of distant pollution sources.

In contrast to the first period, there were few ambiguities concerning the origin of the aerosol of the second period. The trajectories clearly pointed to a Eurasian origin for this pollution event, although at these distances (3000–4000 km) horizontal resolution could not be better than a few hundred km. Confirmation of a Eurasian origin was provided by the noncrustal Mn/noncrustal V ratio of sample 7, the day of haze over Iceland, which was the only day on which both noncrustal Mn and noncrustal V could be detected above the high local background. Rahn (1981b) has shown that the Mn/V ratios of eastern North American and Eurasian aerosols are systemati-

cally different, with Eurasian values 5 ± 1 times higher than those of eastern North America. When proper allowances are made for seasonal variations in this ratio (factors of 2–4 in both North America and Eurasia) and its systematic changes during long-range transport (decreases by factors of 2–3 over the first few thousand km), this ratio can be a powerful indicator of the origin of an aerosol or an air mass. There is now both theoretical and observational evidence that the Mn/V ratio of North American aerosol is 0.2–0.3 after transport to the vicinity of Iceland during summer, whereas Eurasian aerosol near Iceland in August should have and actually has a Mn/V ratio an order of magnitude higher, approximately 2–3 (Rahn, 1981b; Rahn *et al.*, 1981). (The great difference in these ratios is because the greater aging of the North American aerosol, already several times richer in V than is the Eurasian aerosol, enriches the V still further.) By comparison the Mn/V ratio of sample 7 of this study was 4.2 ± 1.9 (Table 1); this value is completely compatible with the Eurasian ratio cited above but not with the North American ratio.

These high Mn/V ratios may be a seasonal effect related to high ratios at the source in summer. Rahn (1981b) has noted that the Mn/V ratios of New York City and central Europe are 3–4 times greater in summer than in winter, which is consistent with the ratios in Iceland during August being 3–6 times higher than they are after transport to the Norwegian Arctic in winter. The ratios in Iceland are also more directly consistent with a Eurasian source: data from the Jungfraujoch (Dams and DeJonge, 1976) can be used to derive a Mn/V ratio of 6–7 for summer. If this aerosol is one-half as aged as is the Icelandic aerosol of sample 7 here, and if transport of a few thousand km decreases the Mn/V ratio by a factor of 2–3 (as it seems to for both Eurasian and North American aerosols: Rahn, 1981b; Rahn *et al.*, 1980; Rahn *et al.*, 1981), the Icelandic Mn/V ratio should be 3.5–5.0. Thus, chemistry and meteorology concur that Eurasia was the source of the aerosol of sample 7.

As noted above, the trajectories showed that the aerosol of samples 5–7 could have been derived from Eurasia. The lower concentrations and lack of visible haze for samples 5–6 and 8–9 suggested that transport for these days was not as efficient or as direct as it was on day 7; this seemed to be confirmed by the $SO_4^=/V$ ratios for samples 5–9. The $SO_4^=/V$ ratio has been shown to be a very sensitive indicator of the degree of aging of a pollution-derived aerosol, because it increases by several-fold during the first few days of transport (Rahn and McCaffrey, 1980; Rahn *et al.*, 1980). $SO_4^=/V$ ratios for this experiment are derived in Table 2. The value for sample 7 was nearly a factor of two lower than that of any of the other four samples of this period, which means that the transport of sample 7 was more rapid and direct than was that of the other samples.

The $SO_4^=/V$ ratios in samples 7–9, which showed the strongest transport from Eurasia, were $1–2 \times 10^3$. We were surprised that these values were so low; on the basis of winter ratios in the Norwegian Arctic ($2–3 \times 10^3$) and the more rapid oxidation of SO_2 in summer, we had expected to find ratios more like the $2–10 \times 10^3$ observed in the Arctic in summer. At present, the explanation for the low Icelandic ratios is not clear. They may imply that dry deposition of SO_2 is considerably greater in summer than in winter, as would seem reasonable in view of the inertness of snow surfaces. Careful examination of data from Bear Island revealed, in agreement with the Iceland situation, that $SO_4^=/V$ of high-concentration episodes during summer is $1–3 \times 10^3$, which is considerably lower than the $2–10 \times 10^3$ for other times. Thus, summer transport of Eurasian aerosol over land or water seems to result in a $SO_4^=/V$ ratio of $1–2 \times 10^3$, comparable to or slightly lower than the $2–3 \times 10^3$ from winter transport. In the future, we will examine the chemometeorological aspects of summer transport in more detail, in the same way as has been done for winter transport by Rahn and McCaffrey (1980) and Rahn *et al.* (1980).

Figure 3 shows that the cloud-active character of aerosol from Europe is markedly different from that of the near-background aerosol of the earlier period. The CCN spectrum from 23 to 24 August, at the height of the transport from Europe, is elevated in concentration and more nearly flat than are the two other spectra. Not only are there more CCN present than earlier, they are more active at lower supersaturations as well. These two features are indicative of continental aerosol, i.e., large numbers of large CCN, as would be expected with transport from Europe. Large CCN are defined here as those active at low supersaturations ($\leq 0.2\%$), regardless of their composition.

Taken together, then, the physical, chemical and meteorological evidence presented here reveals an Icelandic aerosol which was generally near-background in character during summer, except when the air came from Eurasia. A high-concentration event of CCN and pollution aerosol was observed in connection with this Eurasian air, during which the CCN counts were elevated by 2–3 times and the pollution aerosol by up to an order of magnitude.

At this point one may inquire about which constituents of the aerosol contributed most to the CCN. In Fig. 2 there is an obvious similarity between the temporal variations of CCN and $SO_4^=$, particularly during the second period of the experiment. This should come as no surprise, for it has been shown previously that $SO_4^=$ is an important component of CCN and is the most abundant trace constituent of cloud water and precipitation (Junge, 1956; Dinger *et al.*, 1970; Twomey, 1971; Petrenchuk, 1975). $(NH_4)_2SO_4$, NH_4HSO_4, and H_2SO_4 are all highly hygroscopic, and would be expected to readily form haze and cloud droplets at high relative humidities and low supersaturations, respectively. But the CCN and $SO_4^=$ are not perfectly parallel; something else must have contributed CCN.

We have used elementary regression analysis to

determine whether any of the other major constituents of the Icelandic aerosol represented by elements we measured could have contributed significantly to the CCN, according to the equation below:

$$\mathrm{CCN(cm^{-3})} = a + b\ \mathrm{SO_4^=(\mu g\,m^{-3})} + cX \qquad (4)$$

where a is a constant concentration (presumed to be from a constituent not measured here), b is the coefficient for sulfate, X stands for some other constituent of the aerosol, and c is the coefficient of X. The results are shown in Tables 3 and 4. Fitting $SO_4^=$ alone plus the constant (a) to the CCN data explained 75% of the variance. The resulting value of (a) was 61 cm^{-3}. Over the 9 samples, $SO_4^=$ and this unknown constituent contributed roughly equally to the CCN; during the first four samples (the near-background period), the unknown constituent accounted for 85% of the CCN (61 out of 72 cm^{-3}). Interestingly, adding Al (for crustal aerosol), Na (for sea-salt aerosol), or CN (for local pollution aerosol) to the regression changed the results only slightly: a constant component of CCN between 57 and 61 cm^{-3} was always required, the coefficients (b) of $SO_4^=$ ranged between only 0.094 and 0.121, and the extra constituents (crustal aerosol, etc.) contributed less than 10% of the total CCN. (For the case of Na, its coefficient was negative.) Thus, neither the Icelandic crust, nor sea salt, nor local Icelandic pollution seemed to contribute substantially to the CCN: relative concentrations of the constant component, of $SO_4^=$, and of other constituents ranged from 51 to 55%, 40 to 52%, and −6 to 6%, respectively.

The constant component of the CCN may have been NO_3^-, organics, or some secondary marine derivative. As a test of this last possibility, I, a secondary marine derivative, which was more nearly constant during this experiment than anything else that was measured, was regressed along with $SO_4^=$ against the CCN. It went much farther toward explaining the CCN (34% contribution) than did the Al, Na, or CN. But this was probably only because of its near-constancy; its actual concentrations (~1 ng m^{-3}) were far too low to contribute the required numbers of CCN. A more likely source of the constant component was organic material, which is abundant (~1 $\mu g\,m^{-3}$ in marine aerosol) and nearly constant over various oceans (Ketseridis *et al.*, 1976; Duce, 1978).

SUMMARY AND CONCLUSIONS

In summary, then, we have observed a case of strong transport of CCN some 3000–4000 km from Eurasia to Iceland during summer. The CCN increased in parallel with the pollution-derived fraction of the aerosol, particularly with the $SO_4^=$, and may be identified in large measure with it. Thus, the CCN concentration in remote regions may prove to be a very useful indicator of the presence of anthropogenic aerosol. It also can provide a direct measurement of a property that is important climatically.

These results from Iceland also allow us to speculate on CCN in the Arctic. It is now known that pollution aerosol is transported from mid-latitudes to all reaches of the Arctic during winter, and possibly during summer as well (Rahn and McCaffrey, 1979b, 1980; Rahn, 1980). Our measurements in Iceland have shown that CCN are transported over comparable distances from the same source, at least in summer. CCN should thus also be dispersed over the Arctic in winter. According to Fitzgerald and Spyers-Duran (1973) and Hobbs *et al.* (1980), increased CCN from pollution sources shifts the size distributions of cloud droplets toward smaller sizes, with consequent changes in the radiative properties of the clouds. If our measurements of CCN in the Arctic document higher-than-expected concentrations, a similar effect on radiative properties of Arctic clouds, hence on the radiation balance of the Arctic, may be inferred.

Acknowledgements—This work was supported by the Office of Naval Research, Contract N00014-76-C-0435. Special thanks are due to Dr. Flosi Sigurdsson and staff of the Icelandic Meteorological Office, without whose arrangements and cooperation this work could not have been possible. Aerosol samples were analyzed by neutron activation at the Rhode Island Nuclear Science Center.

Table 3. Regression analysis for CCN

		CCN (cm^{-3}) at 0.2% SS				
Sample	Actual	$SO_4^=$ + constant	SO_4 + Al + constant	$SO_4^=$ + Na + constant	SO_4 + CN + constant	SO_4 + I + constant
1	86	80	83	76	85	89
2	92	65	65	61	65	75
3	76	87	84	87	92	86
4	33	83	82	85	80	81
5	74	78	77	78	75	71
6	116	114	120	120	111	105
7	178	202	198	192	203	202
8	160	128	121	129	127	121
9	189	166	173	177	166	176
r^2	-	0.75	0.76	0.77	0.76	0.77

Table 4. Regression coefficients and relative mean contributions of several possible contributors to the CCN over Iceland

Component of CCN	Mean value	Regression coefficient					Relative mean contribution to CCN, %				
Nonmarine $SO_4^=$ (secondary nonmarine)	480 ng m^{-3}	0.105	0.094	0.121	0.105	0.065	45	40	52	45	28
Al (crust)	409 ng m^{-3}		0.017					6			
Noncrustal Na (primary marine)	412 ng m^{-3}			−0.017					−6		
CN (local contam.)	730 cm^{-3}				0.0054					4	
1 (secondary marine)	1.06 ng m^{-3}					36.17					34
Constant component	cm^{-3}	61.1	59.8	60.2	57.1	41.9	55	53	54	51	38
CCN	112 cm^{-3}										

REFERENCES

Barrie L. A., Hoff R. M. and Daggupaty S. M. (1981) The influence of midlatitudinal pollution sources on haze in the Canadian Arctic. *Atmospheric Environment* **15**, 1407–1419.

Blanchard D. C. (1969) The oceanic production rate of cloud nuclei. *J. rech. Atmos.* **4**, 1–6.

Borys R. D. (1980) A modified Mee Industries static, thermal gradient, diffusion cloud chamber. *Proc. Int. CCN Workshop*, 6–17 October 1980, Desert Research Institute, University of Nevada, Reno, NV.

Dams R. and DeJonge J. (1976) Chemical composition of Swiss aerosols from the Jungfraujoch. *Atmospheric Environment* **10**, 1079–1084.

Dinger J. E., Howell H. B. and Wojciechowski T. A. (1970) On the source and composition of cloud nuclei in a subsident air mass over the North Atlantic. *J. atmos. Sci.* **27**, 791–797.

Duce R. A. (1978) Speculations on the budget of particulate and vapor phase nonmethane organic carbon in the global troposphere. *Pageoph.* **116**, 244–273.

Elliott W. P. and Egami R. (1975) CCN measurements over the ocean. *J. atmos. Sci.* **32**, 371–374.

Fitzgerald J. W. and Spyers-Duran D. A. (1973) Changes in cloud nucleus concentration and cloud droplet size distribution associated with pollution from St. Louis. *J. appl. Met.* **12**, 511–516.

Fletcher J. O. (1965) The heat budget of the Arctic Basin and its relation to climate. Report R-444-PR, The RAND Corporation, 1700 Main St., Santa Monica, CA 90406, 179 pp.

Gjessing Y. T. (1977) Episodic variations of snow contamination of an Arctic snowfield. *Atmospheric Environment* **11**, 643–647.

Gmelin (1968) *Gmelin's Handbuch der Anorganischen Chemie*. Achte Auflage: Vanadium, Teil A. Lieferung 1. System Nummer *48*, 43, 48 u. 49. Weinheim: Verlag Chemie. 1968.

Gravenhorst G. (1978) Maritime sulfate over the North Atlantic. *Atmospheric Environment* **12**, 707–713.

Hobbs P. V. (1971) Simultaneous airborne measurements of cloud condensation nuclei and sodium-containing particles over the ocean. *Q. Jl R. met. Soc* **97**, 263–271.

Hobbs P. V., Stith J. L. and Radke L. F. (1980) Cloud-active nuclei from coal-fired electric power plants and their interactions with clouds. *J. appl. Met.* **19**, 439–451.

Hoppel W. A. (1979) Measurement of the size distribution and CCN supersaturation spectrum of submicron aerosols over the ocean. *J. atmos. Sci.* **36**, 2006–2015.

Jayaweera K. O. L. F. and Ohtake T. (1973) Concentration of ice crystals in Arctic stratus clouds. *J. rech. Atmos.* **8**, 199–207.

Jiusto J. E. (1966) Maritime concentrations of condensation nuclei. *J. rech. Atmos.* **1**, 245–250.

Junge C. E. (1956) Recent investigations in air chemistry. *Tellus* **8**, 127–139.

Ketseridis G., Hahn J., Jaenicke R. and Junge C. (1976) The organic constituents of atmospheric particulate matter. *Atmospheric Environment* **10**, 603–610.

Larssen S. and Hanssen J. E. (1980) Annual variation and origin of aerosol components in the Norwegian Arctic-Subarctic region. In *Proceedings of the WMO Technical Conference on Regional and Global Observation of Atmospheric Pollution Relative to Climate*, Boulder, CO, 20–24 August 1979.

Mason B. (1966) *Principles of Geochemistry*. 3rd ed. John Wiley, New York.

Ottar B. (1980) The long range transport of sulphurous aerosol to Scandinavia. *Ann. N.Y. Acad. Sci.* **338**, 504–514.

Peterson J. T., Hanson K. J., Bodhaine B. A. and Oltmans S. J. (1980) Dependence of CO_2, aerosol, and ozone concentrations on wind direction at Barrow, Alaska

during winter. *Geophys. Res. Lett.* **7**, 349–352.

Petrenchuk O. D. (1975) Estimation of condensation nuclei chemical composition from the results of cloud water analysis. *Proceedings of the 8th International Conference on Nucleation*, Leningrad, USSR, 24–29 September 1973.

Pytkowicz R. M. and Kester D. R. (1971) The physical chemistry of sea water. *Oceanogr. Mar. Biol. A. Rev.* **9**, 11–60.

Radke L. F., Hobbs P. V. and Pinnons J. E. (1976) Observations of cloud condensation nuclei, sodium-containing particles, ice nuclei and the light-scattering coefficient near Barrow, Alaska. *J. appl. Met.* **15**, 982–995.

Rahn K. A. (1980) On the causes, characteristics and potential environmental effects of aerosol in the Arctic atmosphere. *Proceedings of the Conference on the Arctic Ocean*, London, 11–12 March 1980 (in press). Sponsored by the Arctic Committee of Monaco and the Royal Geographic Society.

Rahn K. A. (1981a) Relative importances of North America and Eurasia as sources of Arctic aerosol. *Atmospheric Environment* **15**, 1447–1455.

Rahn K. A. (1981b) The Mn/V ratio as a tracer of large-scale sources of pollution aerosol for the Arctic. *Atmospheric Environment* **15**, 1457–1464.

Rahn K. A. (1981c) Atmospheric, riverine and oceanic sources of seven trace constituents to the Arctic Ocean. *Atmospheric Environment* **15**, 1507–1516.

Rahn K. A., Borys R. D. and Shaw G. E. (1977) The Asian source of Arctic haze bands. *Nature* **268**, 713–715.

Rahn K. A., Brosset C., Ottar B. and Patterson E. M. (1980) Black and white episodes, chemical evolution of Eurasian air masses, and long-range transport of carbon to the Arctic. *Proceedings of the General Motors Research Symposium Particulate Carbon: Atmospheric Life Cycle*, 12–14 October 1980, Warren, Michigan (in press).

Rahn K. A., Conway T. J. and Ray B. J. (1981) North American and Eurasian aerosol between Halifax and Reykjavik during August 1971 (in preparation).

Rahn K. A. and McCaffrey R. J. (1979a) Compositional differences between Arctic aerosol and snow. *Nature* **280**, 479–480.

Rahn K. A. and McCaffrey R. J. (1979b) Long-range transport of pollution aerosol to the Arctic: A problem without borders. *Papers presented at the WMO Symposium on the Long-Range Transport of Pollutants and its Relation to the General Circulation including Stratospheric/Tropospheric Exchange Processes*, Sofia, 1–5 October 1979, WMO-No. 538, pp. 25–35.

Rahn K. A. and McCaffrey R. J. (1980) On the origin and transport of the winter Arctic aerosol. *Ann. N.Y. Acad. Sci.* **338**, 486–503.

Shaw G. E. and Stamnes K. (1980) Arctic haze: Perturbation of the polar radiation budget. *Ann. N.Y. Acad. Sci.* **338**, 533–539.

Stevenson C. M. (1968) An improved Millipore filter technique for measuring the concentrations of freezing nuclei in the atmosphere. *Q. Jl R. Met. Soc.* **94**, 35–43.

Sze N. D. and Ko M. K. W. (1980) Photochemistry of COS, CS_2, CH_3SCH_3 and H_2S: Implications for the atmospheric sulfur cycle. *Atmospheric Environment* **14**, 1223–1239.

Tabatabai M. A. (1974) A rapid method for determination of sulphate in water samples. *Envir. Lett.* **7**, 237–243.

Twomey S. (1959) The nuclei of natural cloud formation II. The supersaturation in natural clouds and the variation of cloud droplet concentrations. *Geofis. Pura Appl.* **43**, 243–249.

Twomey S. (1971) The composition of cloud nuclei. *J. atmos. Sci.* **28**, 377–381.

Twomey S. and Wojciechowski T. A. (1969) Observations of the geographical variation of cloud nuclei. *J. atmos. Sci.* **26**, 684–688.

Vinogradov A. P. (1959) *The Geochemistry of Rare and Dispersed Chemical Elements in Soils*, 2nd ed., Consultants Bureau, Inc., New York.

Weiss H. V., Herron M. M. and Langway C. C. Jr. (1978) Natural enrichment of elements in snow. *Nature* **274**, 352–353.

Zoller W. H., Gordon G. E., Gladney E. S. and Jones A. G. (1973) The sources and distribution of vanadium in the atmosphere. In: *Trace Elements in the Environment*, Advances in Chemistry Series, No. 123, *Am. Chem. Soc.*, Washington, DC, pp. 31–47.

APPENDIX D

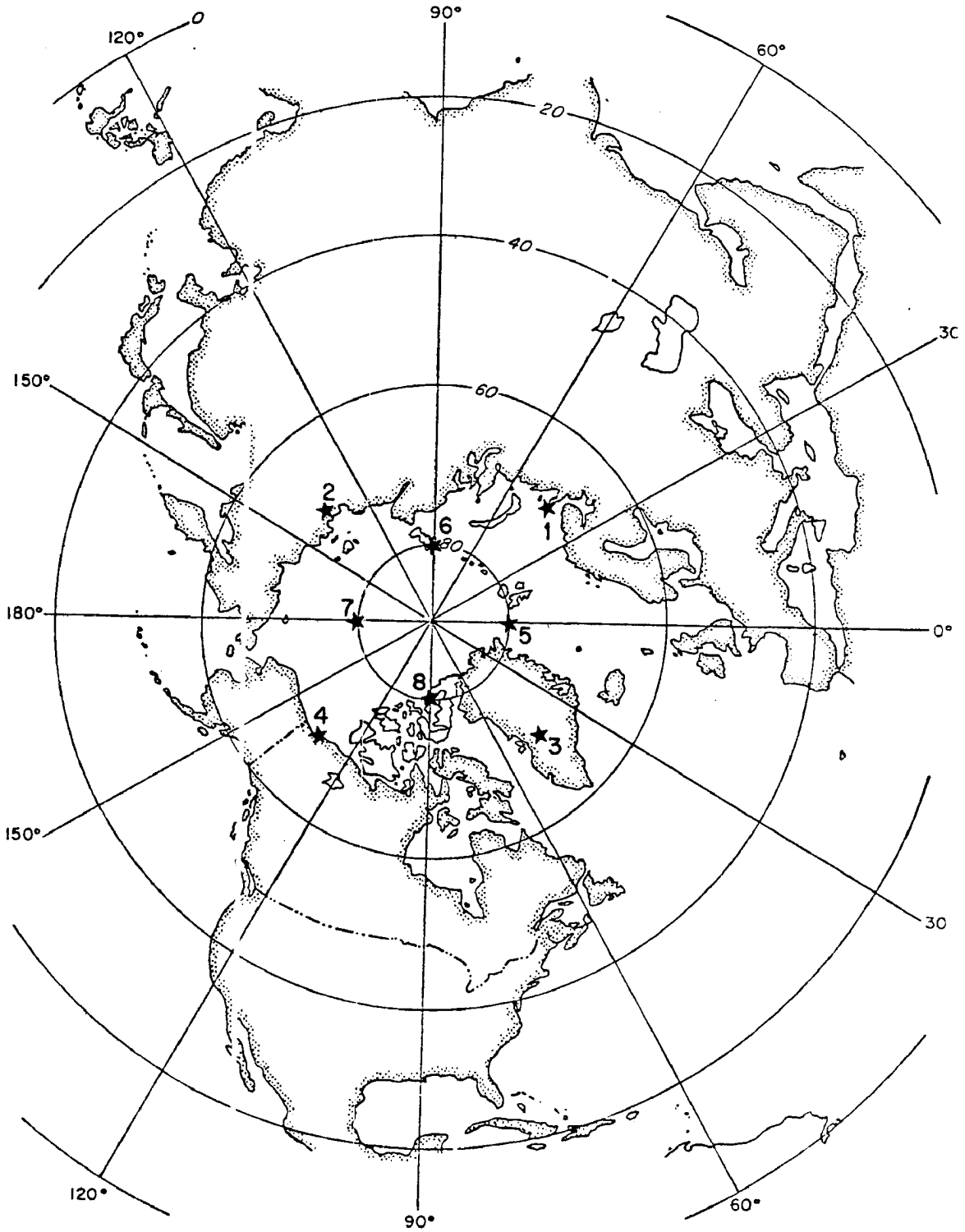


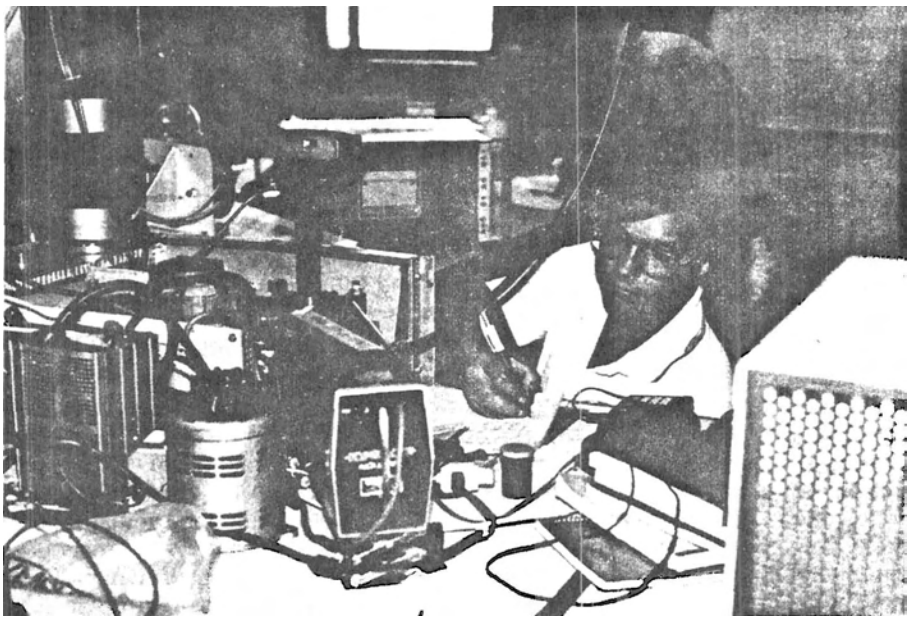
Figure 1: Locations of the eight grid points chosen along the 70 N and 80 N latitude circles. Geostrophic wind speed and direction were determined at each of these points at 500 mb.

J. Rech. Atmos., 15 (1981), n° 3-4, pp. 267-269.
THIRD INTERNATIONAL CLOUD CONDENSATION NUCLEI WORKSHOP.
Section V. Instrument Descriptions.

MODIFIED MEE INDUSTRIES STATIC THERMAL GRADIENT DIFFUSION CLOUD CHAMBER

by

Randolph D. BORYS
Department of Atmospheric Science
Colorado State University
Fort Collins, Colorado, U.S.A.



1. — EQUIPMENT DESCRIPTION.

The purpose of attending the CCN Workshop was to calibrate and determine the usable operating characteristics of the cloud chamber and detection system described below. The instrument consisted of a Mee Model 130 Cloud Condensation Nucleus Counter (Serial # 3). The original optical bench, light source, and detection components were removed and replaced with a simple laser illumination-photographic counting system. The purpose of the modification was to permit discrete droplet counting rather than the original method of scattered light detection and to extend the minimum concentration detection limit to approximately 10/cm^3 for use in remote areas where aerosol concentrations were expected to be low. The system consisted of a 5 mW HeNe laser with an operating wavelength of 0.65 microns. The laser beam was incident to the camera field of view at a 40° angle, the same angle used in the original instrument

configuration utilizing a photo diode as a scattered light detector. In both arrangements, the detector, in this case the camera, sensed the forward scattered light at the given angle from the center of the parallel plate thermal gradient diffusion chamber. The film used was Kodak 2475 recording film developed at 3000 ASA. Photographs were made at 1/2 second exposure through a microscope attachment to the chamber producing a magnification of 3.47 × on the film negative. The observed volume was 0.011 cm^3. Temperature and upper and lower plate ΔT were controlled by thermoelectric cooling of the bottom plate with the upper plate maintained at ambient temperature.

The operation procedure consists of flushing the chamber for 10 seconds at a sample flow rate of 4 liters per minute. An observation is then made through the eyepiece to determine the time required for maximum droplet number concentration to occur. This time is then used for subsequent measurements at the same selected chamber conditions. 1 to 30 exposures are made on each frame to obtain 50-100 droplet images per frame, depending upon the CCN concentration. This produces uncertainties of 10-15 % in the CCN concentration determination. At the upper limit of 30 exposures used, concentrations of 10 cm^{-3} can be determined with uncertainties of 50 %. The time to determine a four-point CCN spectrum of low concentrations (< 500 cm^{-3} at 0.5 % SS) is 90 minutes. At high concentrations the time reduces to about 30 minutes. Since the droplet counting is from images of droplets on film, the minimum detectable concentration and uncertainty can be selected by the operator with the subsequent loss or gain of time resolution. The instrument in its present form can only be operated manually.

2. — RESULTS OF THE WORKSHOP.

The experiments performed at the Workshop were undertaken to confirm the calculated sample volume, determine the usable range of supersaturation, and minimum detectable droplet size.

Problems with chamber leaks were discovered for the experiments 0-10. Hence the data from these experiments cannot be used for comparison.

Comparison of CCN concentrations with other state-of-the-art continuous flow diffusion chambers at supersaturations near 1 % indicated that the calculated sample volume used produced CCN concentrations well within the range of CCN concentrations determined at the Workshop. This agreement is interpreted to mean the sample volume was correct. Direct measurements of beam diameter conducted in the laboratory would have produced errors in CCN concentrations by a factor of 4-5 too low. This error is apparently due to the larger apparent visible beam diameter versus the actual usable beam diameter given the droplet illumination, chamber optical geometry, microscope optics and film characteristics. It is therefore suggested that the only way in which the sample volume should be determined is by experimentation with known droplet sizes and numbers via known aerosol generation and sampling.

By comparison of the results from experiments 19 and 15, it was determined that the minimum usable supersaturation for this chamber was between 0.09 % and 0.22 %, respectively. By using the dry aerosol data from experiments 15, 18, 19 and 20, and the number of droplets detected at indicated supersaturation of 0.1 %, the minimum supersaturation the instrument was able to achieve was 0.11 %, 0.17 %, 0.09 %, and 0.19 %, respectively. Therefore, 0.20 % was determined as the lowest usable supersaturation.

A minimum detectable size of about one micron radius was determined, based upon the smallest observed image on the film, the known magnification of the image by the microscope attachment, and the artificial enlarging of the image by diffraction of the relatively long wavelength monochromatic light through the small diameter lenses of the microscope. This agrees roughly with a droplet size of 0.7 micron radius at which the maximum in light scattering efficiency occurs for the wavelength light used.

The chamber response to known aerosol at different supersaturations compared favorable to theory, both in the predicted slope for polydisperse aerosol and the determination of the critical supersaturation for monodisperse aerosol. However, there appears to be some tendency to undercount at mid-range supersaturations giving an anomalous concave upward appearance to the curve for some experiments, but not all. The cause of this effect is unknown. Further testing will be required to look more closely at this effect. Table 1 summarizes the pertinent instrument parameters which existed for the instrument as it was operated during the Workshop.

TABLE 1

Instrument parameters

Detection System :	
Laser light source	5 milliwatts 0.65 micron wavelength
Photographic counting	2475 recording film ASA 3000
Minimum detection size	$\sim$ 1 μm radius
Sampling :	
Flow rate	4 lpm for 10 sec
Sample volume	0.011 cm^3
Time for spectra (4 pts)	30-90 min
Chamber Specifications :	
Aspect ratio	4.6:1
Temperature control	Thermoelectric cooling of lower plate
Usable supersaturation range	0.2 – 2.0 %

ACKNOWLEDGEMENTS.

The authors participation was partially sponsored by ONR Contract N00014-79-C-0793.

APPENDIX E

DETAILED FIELD EXPERIMENT DISCUSSION
Narragansett, Rhode Island, USA

This site represents a mid-latitude source region with respect to the aerosol particle's physical and chemical properties. Located essentially on the coast of the Atlantic Ocean between Boston, Massachusetts and New York, New York, it is within the east coast urban corridor. It is locally situated in a rural environment without any heavy industry nearby. This site can be under the influence of different air mass and aerosol-particle source regions depending upon the wind direction. This is apparent when the aerosol particle composition and the meteorology are compared.

E.1 Summer Study

The summer aerosol study at this site was conducted during the period 12-19 June, 1979. Seven, 24-hour samples were collected and analyzed. The bulk aerosol particle trace element chemistry is given in Table 1, Appendix A. Figure E.1 is a plot of the day-to-day variation of the aerosol source tracer components of the total aerosol defined earlier and the CCN at 0.2% SS and the IN at $-20^{o}C$ at 1% water SS. Figures E.2a-d are isohypse analyses for the period for the 850 mb and 700 mb surfaces valid at 0Z. Each isohypse has up to three hatch marks corresponding to the distance of travel back in time along the isohypse using the wind speed observed at the proper level over the site. This approximates the speed of transport at the time of observation. The

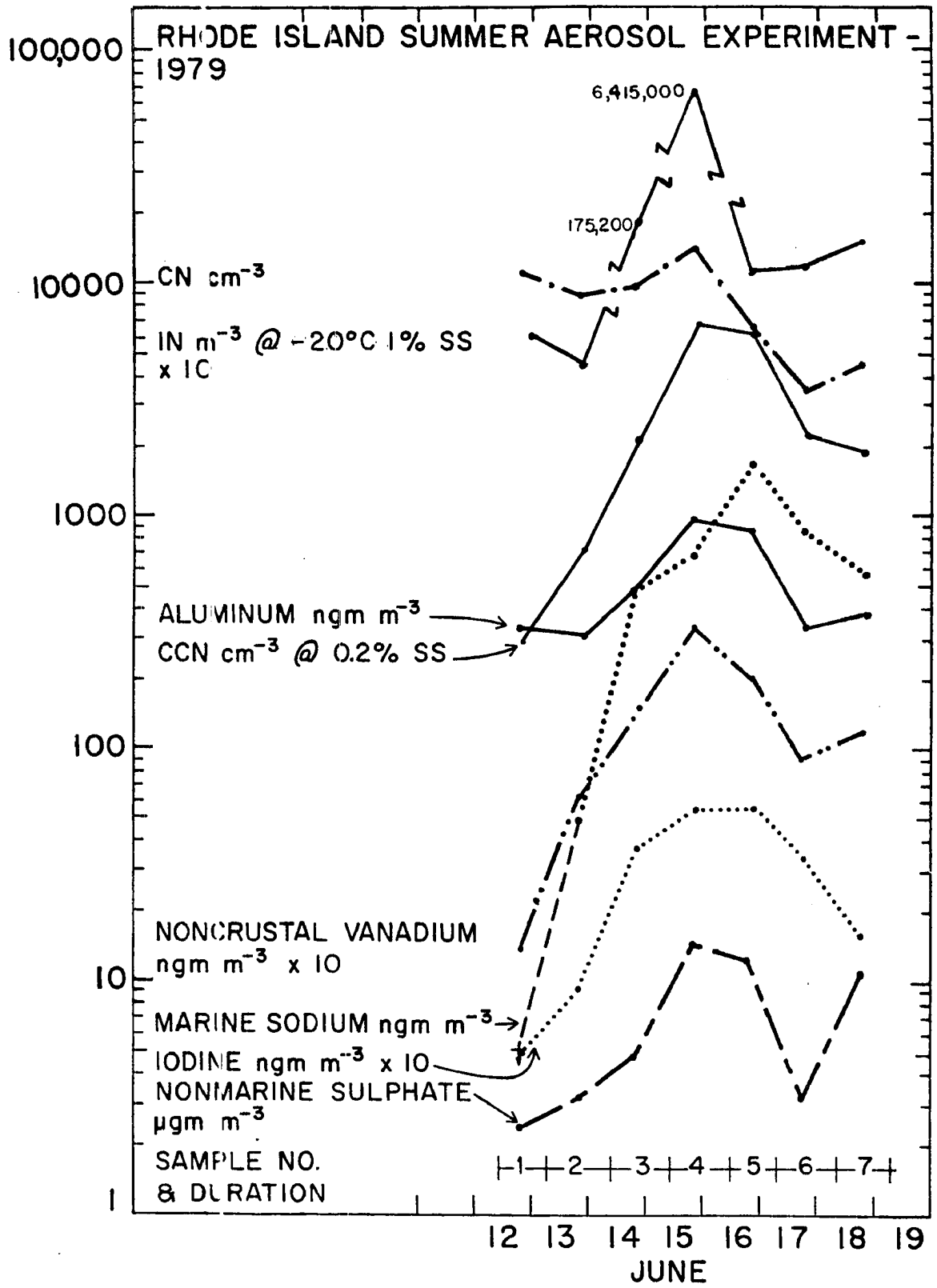


Fig. E.1 Narragansett, Rhode Island, USA summer aerosol experiment, June 1979.

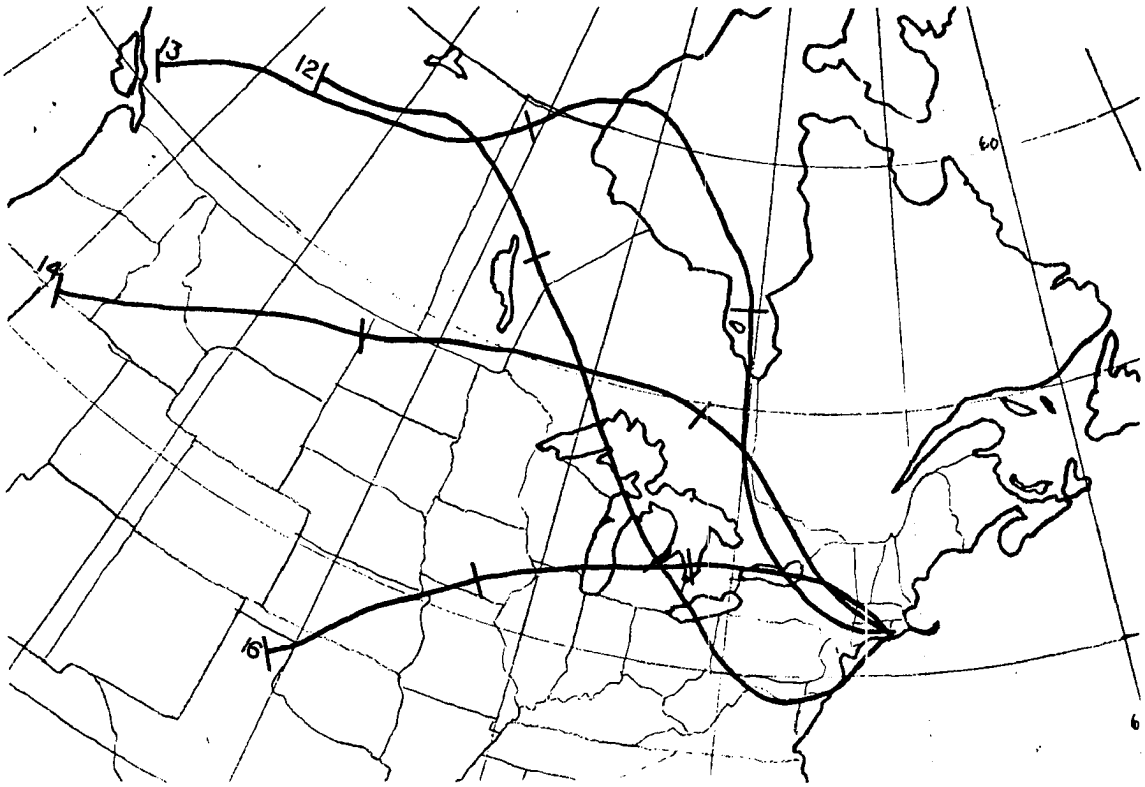


Fig. E.2b Same as a at 700 mb.

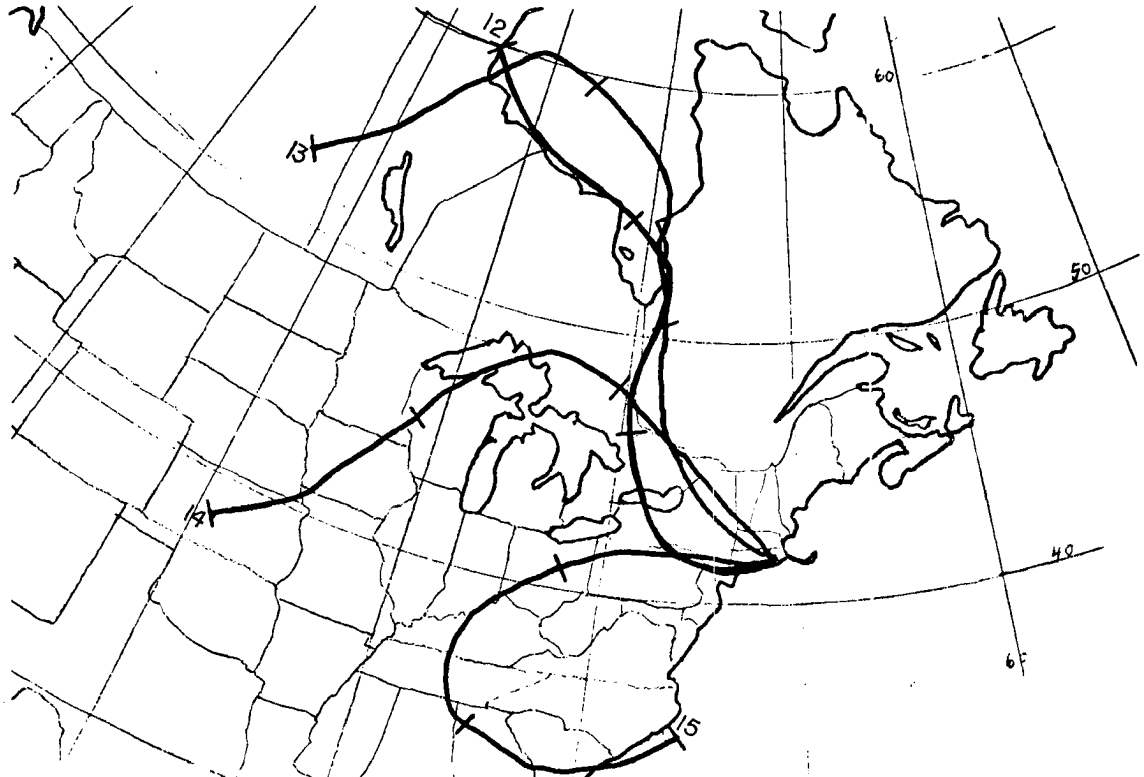


Fig. E.2a Narragansett 850 mb isohypse analyses, 12 - 15 June, 1979, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

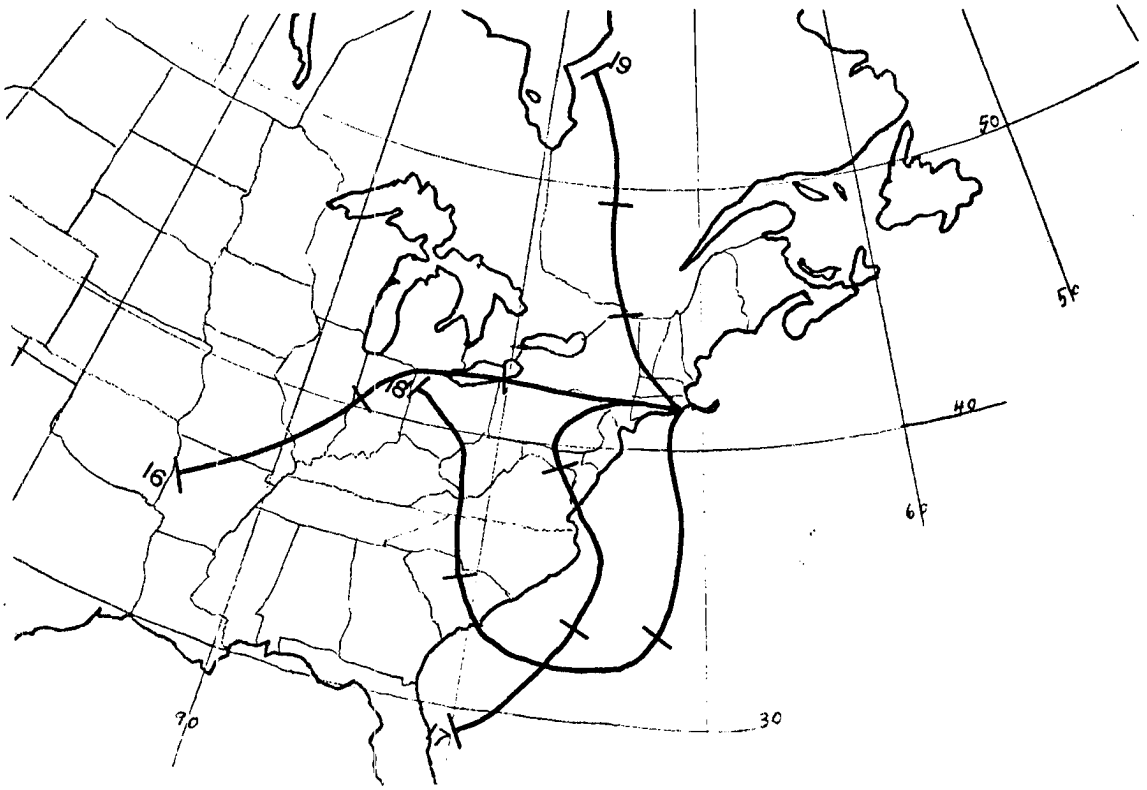


Fig. E.2d Same as a at 700 mb, 16 - 19 June, 1979.

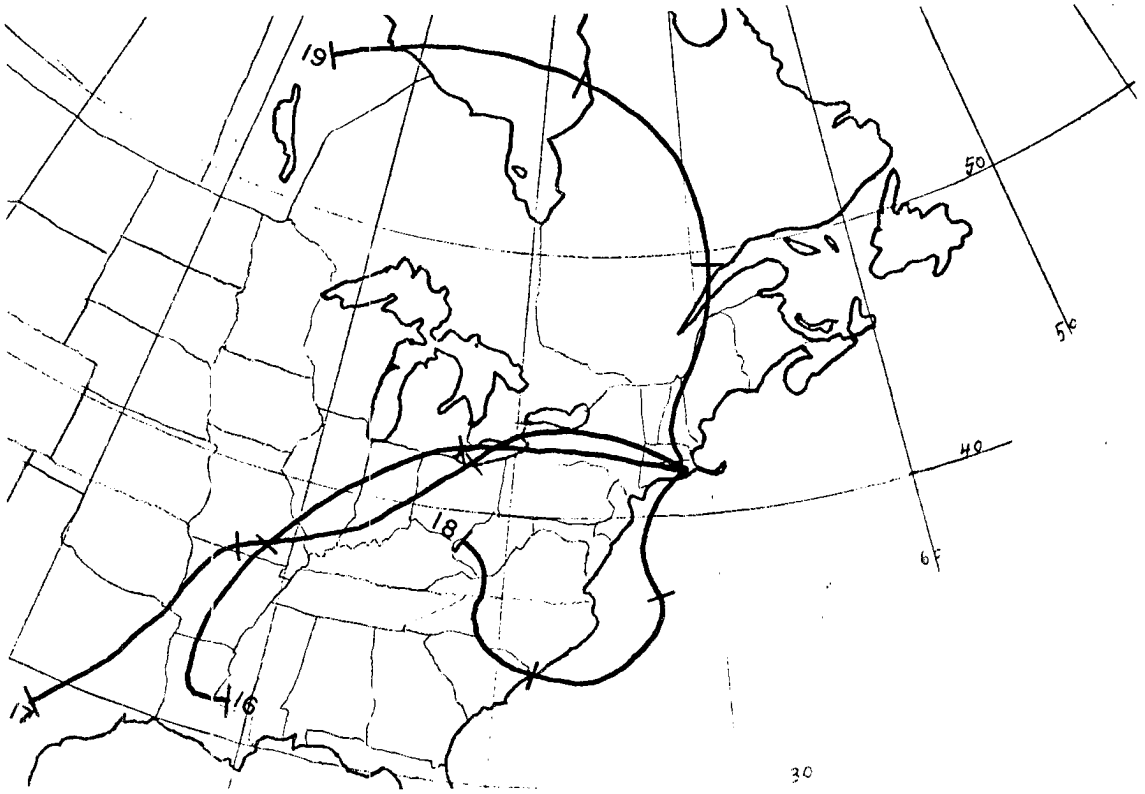


Fig. E.2c Same as a at 850 mb, 16 - 19 June, 1979.

number at the end of each isohypse corresponds to the date for which the isohypse is valid.

The measured aerosol particle trace element chemistry is typical for this site. Since it is mid-June, the high loadings associated with midsummer conditions are not present. The period of study encompassed a short wave passage. This emerges from the aerosol chemistry data of Fig. E.1. Initially the winds were from the north to west which advected air from Canada to the site. Winds from this direction bring cool, dry continental air, relatively low in pollutant aerosol concentration because of few major upwind sources. With time, the wind direction began to back through the west to south as a trough approached. This gradually advected air from more polluted source regions. This finally placed the New York City region upwind of the sampling site. This gradual progression from a "background" continental air mass to a polluted urban air mass can be seen in the change in the aerosol trace element tracers. As expected, the overall increase in aerosol loading is reflected in all of the elemental tracers. Southerly winds brought more of a maritime airmass to the sampling location on June 17 and 18. This gradually displaced the pollutant aerosol particles. This is evident in the peaking of the marine sodium (Na) just after the maxima in non-marine sulphate ($SO_4^=$) and non-crustal vanadium (V). By sample 6 the aerosol particles were maritime. The final sample begins to show the return to a more continental aerosol composition as the short wave passes out to sea and the winds come back to a northwesterly direction.

This sample set provides a unique opportunity to relate the observed changes in airmass and aerosol composition to the concentration

of the cloud active aerosol. In this situation where the pollution sources are relatively nearby and dominate the total aerosol, the pollutant aerosol is the major source of IN and CCN at this site. The maxima in both CCN and IN occur on sample 4, the sample with the heaviest pollutant aerosol tracer concentration. The maximum IN concentration (6.4×10^6 m^{-3}) and CCN concentration (6.4×10^3 cm^{-3}) are the highest measured during this project. These results are consistent with the observations of others in the urban plumes are strong sources of CCN. The results of others for IN pollutant sources may not be as clear. However, two comments are warranted here. 1) The measurements made here and at the other arctic sites in this study for IN are most useful when used for comparison within a sample set from a given site and season. Thus, in this case the relative importance of the urban aerosol source region is much larger than that of the maritime and continental regions for this location. 2) In this particular case, the high concentrations of IN are closely related to the high concentrations of CCN. This is a result of the correction factor used for the IN concentations as given by Huffman and Vali (1973). The correction factors for samples 3 and 4 are 2800 and 134,200 respectively. This method of correction may not be applicable in situations of high aerosol particle concentrations. However, the correction has a sound physical basis. For this measurement technique, the results should be used for qualitative relative interpretation within the sample set. The IN results at the other temperatures of -16^{o}, -24^{o}, and -28^{o} are all qualitatively the same. The results are considered to be representative of IN and CCN concentrations in a relatively fresh (<1 day old) urband aerosol plume.

Nuclepore filter samples of the aerosol were also collected for study using a scanning electron microscope (SEM). Samples 1 and 4 were photographed at magnifications of 3100X and 10,000X to compare the morphology of the two distinctly different aerosol populations. The area in the 3100X pictures represents approximately 40–50 cm^3 of sampled aerosol. The 10,000X pictures represent 4–5 cm^3 of sample volume. This is true in general for the SEM photographs at all sites. The photographs are presented in Figures E.3a-d. Comparison of the observed particle loadings to the elemental analysis for the two samples shows an apparent discrepancy. While most trace elements increase in concentration by approximately an order of magnitude from sample 1 to 4, the apparent loading on filter 4 is less than that of 1 from comparison of the photographs. Close inspection of the filter from sample 1 reveals the filter to be covered with many dark appearing, flat particles. In addition, many of the 0.2 μm diameter pores of the filter have been filled with a substance. Detailed elemental analysis by x-ray emission using the SEM electron microprobe of these flat, platelike particles only indicated the presence of sulphur. Both this analysis and the elemental analysis used on the bulk aerosol particles are not sensitive to organic materials comprised mainly of carbon, hydrogen and nitrogen. The concentrated electron beam used to analyze the particles in the SEM vaporized the particles. This suggests the particles are composed of a volatile substance, probably organic and containing sulphur. It is not known whether these particles are artifacts of the sampling procedure or actual collected ambient aerosol particles.

The trace element analysis is a method which utilizes known trace elemental composition of major natural and anthropogenic aerosol

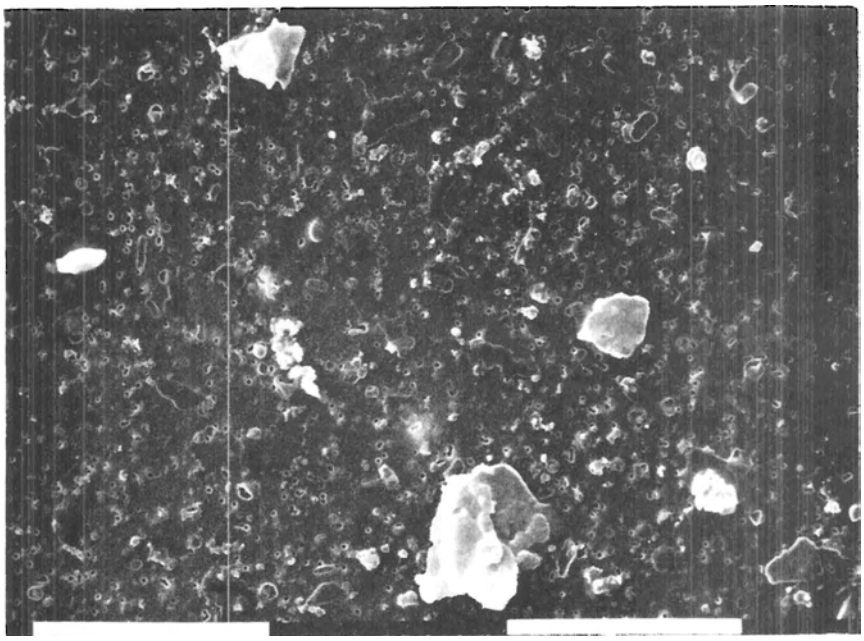


Fig. E.3a Scanning electron microscope photographs of aerosol particles collected at Narragansett during summer on 0.2 μm diameter pore size Nuclepore filters from sample #1, 3100X magnification, bar scale is 10 μm.

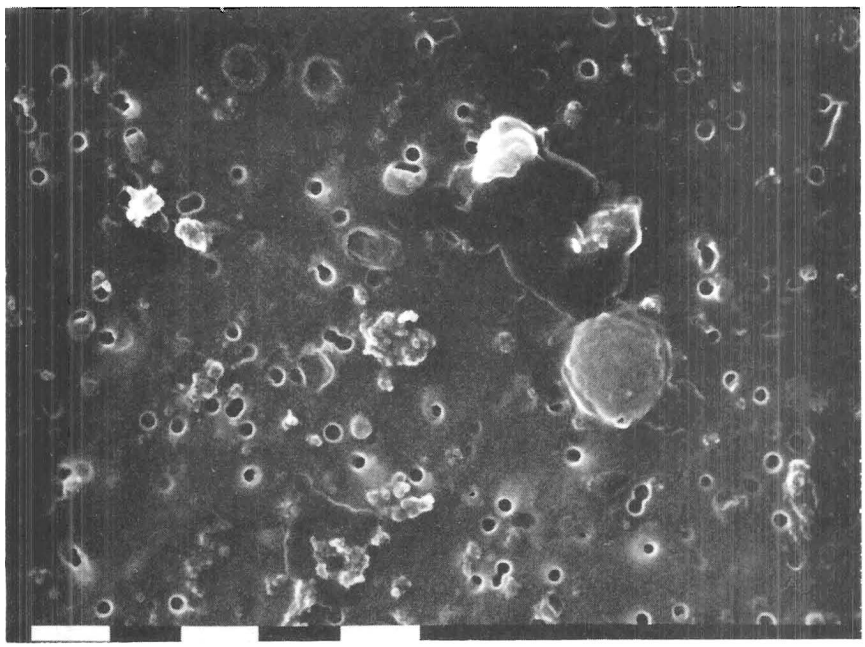


Fig. E.3b Same as a, but 10,000X magnification, bar scale is 1 μm.

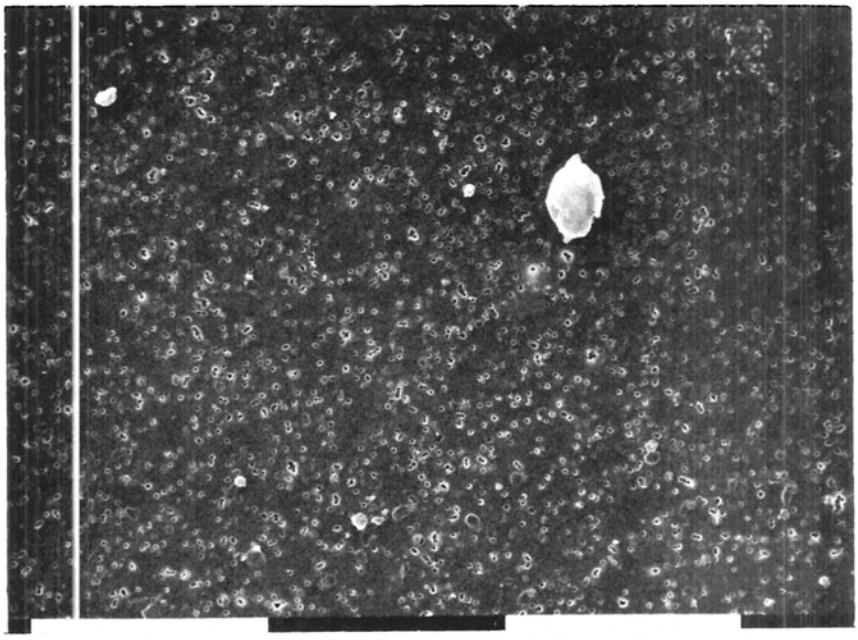


Fig. E.3c Same as a but for sample #4.

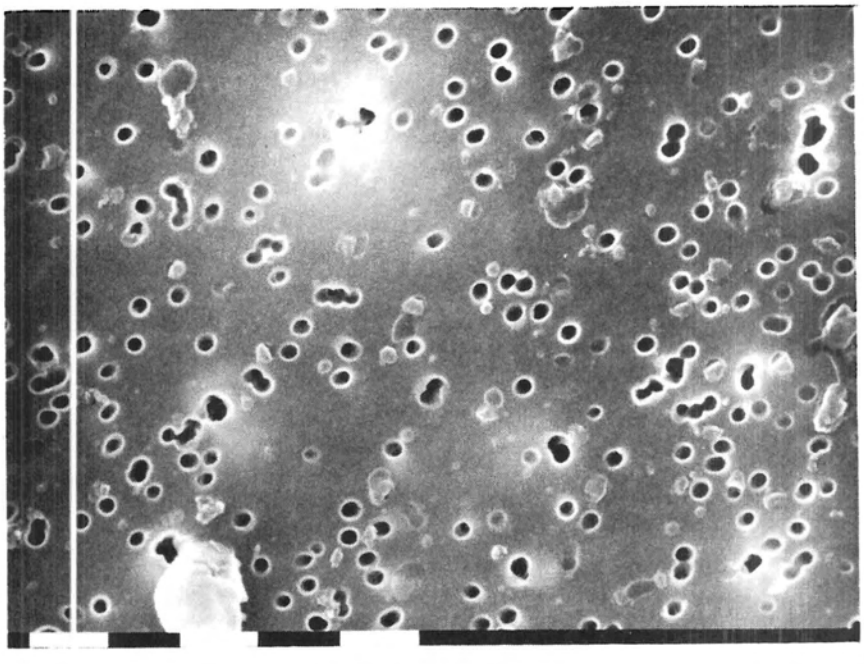


Fig. E.3d Same as c, but 10,000X magnification, bar scale is 1 μm.

particle sources. It may not reflect changes in the organic aerosol particle component as these two photographs imply. This organic component may be a major fraction of the total aerosol mass. It is unknown what effects this material may have on the cloud active aerosol. Since little is known about organic CCN or IN, no attempt will be made to ascribe any observed CCN and IN concentrations or activity to the organic component of the aerosol. It is important to be aware that this component exists. The well known elemental tracer techniques will be used to ascribe the observed variations of the cloud active aerosol to the aerosol sources which the elements represent.

A linear correlation matrix was constructed from the aerosol particle tracer element chemistry and IN and CCN data. These data are presented in Table 5.1. All data from all sites are summarized in this table. Listed by site and season is the per cent variance explained of the IN and CCN concentrations by the various aerosol particle chemical tracers. Values which are underlined indicate significance at the 95% level. Negative values indicate a negative correlation. For the Narragansett summer aerosol experiment the results reflect the dominating role of the total aerosol loading on the relationship of the cloud active aerosol to the aerosol source.

E.1.a Cloud condensation nuclei

The CCN correlate strongly and positively with all the tracers except CN. This indicates that fresh aerosol particles, which CN concentrations represent from the many combustion sources near the site are not important as sources of CCN. This is in line with observations by Twomey and Severynse (1964) of CCN and CN on Long Island, New York where CCN at 1% supersaturation only represented 0.4-2.3% of the total

CN concentration. Strong positive correlation occurs with the tracers indicating the presence of pollution sources ($SO^{=}_4$ and V) and a continental (Al) airmass. However, it would be difficult in this case to separate pollution sources of Al (such as flyash) from the natural background. The SEM photograph of sample 4 in Figure E.3c and d shows the presence of many, small ($\approx$ 0.1 μm radius) which appear to be droplet residues. This is in line with the observed CCN. The ssecondary marine aerosol particle tracer (I) also has a high variance value. This results from the fact that the major transport of pollutant aerosol to Narragansett from the New York City area occurs when the winds are southwest. This pathway lies along the coastline, is primarily associated with low wind speeds and is associated with warm, nutrient rich, biogenically productive marine waters which are suspected to be the source of the iodine vapor precursors to the iodine aerosol particles from marine areas. An interesting result is the relatively low value of the variance for non-crustal Na. This is an indicator of primary marine aerosol particles, predominantly sea spray. Coastal areas are major sources of sea spray yet the pollutant aerosol component appears to dominate the maritime primary aerosol particle component as a source of CCN.

Overall, because of the close proximity of a major pollution source region, it is difficult to assess the relative importance of the natural aerosol on the CCN concentrations. The data shows that the pollution aerosol is a major component, with the marine and continental aerosol playing a lesser role. It is possible to make a first estimate of the relative importance to the total CCN concentration of the natural continental and marine sources versus the pollutant sources. The smooth

transition of wind direction from north through west to south with the concurrent changes of the aerosol particle tracer composition with time allows the source regions to be assigned to different samples. From the meteorology and chemistry, it is apparent that sample 1 is representative of a continental source for this region. Sample 4 represents a combination of pollutant, continental and marine components. The most marine-like sample, 6, has too high of a CCN concentration. This is most likely a result of transport of a polluted continental airmass off shore along the southside of the low pressure center moving eastward and its return to shore as a modified marine-pollution airmass. Let a typical value for CCN active at 0.2% supersaturation over the ocean be 100 cm^{-3}. Let sample 1 with a CCN concentration at the same supersaturation of 280 cm^{-3} represent the background continental CCN concentration. Summing these values, and subtracting from the CCN concentration of sample 4 leaves a residual of 6120 cm^{-3}. Thus in this location, the presence of a polluted airmass can be responsible for up to 94% of the total CCN. This is in line with the overall increase in the total aerosol mass loading which accompanies the polluted airmass.

E.1.b Ice nuclei

As previously discussed, the interpretation of the IN concentrations is dependent upon the validity of the IN concentration correction factor under large aerosol mass loadings. Assuming that it is valid, the results in Table 5.1 show that the IN concentration is strongly and positively correlated with the non-crustal V, the primary pollutant aerosol particle tracer. The per cent variance explained by Al is also high. These two components of the aerosol particle

population are associated with insoluble particles. This a a prerequisite for a particle with ice nucleating ability. The soluble components of the aerosol, marine Na and non-marine $SO^{=}_4$, and the marine secondary organic component tracer, I, have little to no covariance with the IN concentration. Based upon these results, the IN concentrations at this mid-latitude source region site are related to the pollutant emissions from major sources nearby. The exact nature of this relationship cannot be deduced from this limited data set. The increase in IN concentrations as the result of pollutant advection to the site may only be a result of the dominance of the increase in aerosol particle numbers and mass over that of the background aerosol providing more potential sites for ice nucleation, rather than an aerosol population with more efficient ice nuclei. Further studies of this type over longer periods of time would be required to further clarify the situation.

E.2 Winter Study

The winter aerosol study at this site was conducted during the period 6-16 January, 1980. Ten 24 hour samples were collected. The bulk aerosol particle trace element chemistry is given in Table 2, Appendix A. Figure E.4 is a plot of the day-to-day variaon of the aerosol source tracer components of the total aerosol collected and the concentrations of CCN at 0.2% SS and IN at -20^{o}C and 1% water SS. Figures E.5a-d are the plotted isohypse analyses for the period of the study for the 850 mb and 700 mb pressure surfaces valid at 0Z. Each hatch mark represents one day movement at observed wind speeds. SEM photographs of collected aerosol particles for samples 3 and 6 are presented in Figures E.6a-d.

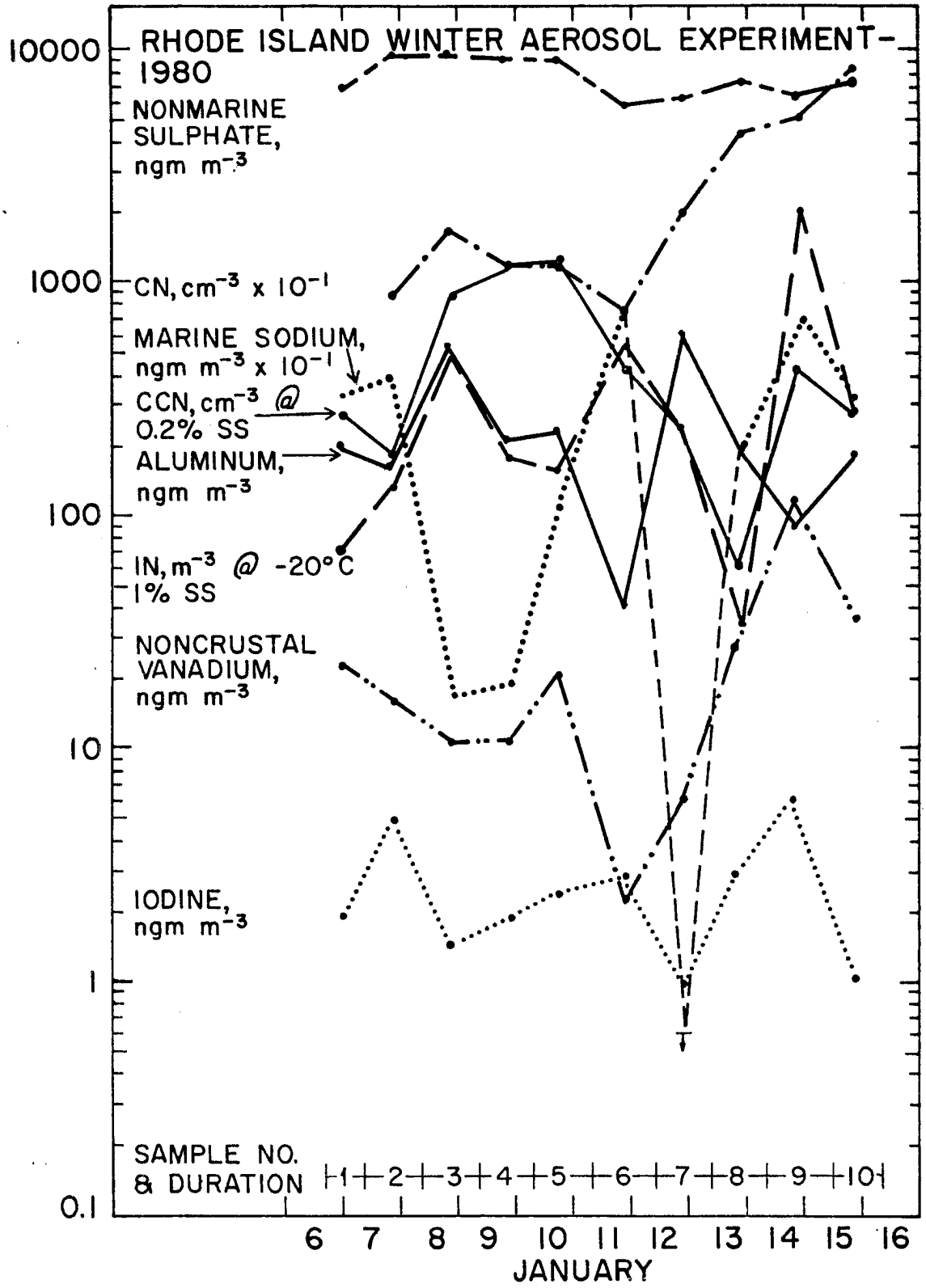


Fig. E.4 Narragansett, Rhode Island, USA winter aerosol experiment, January 1980.

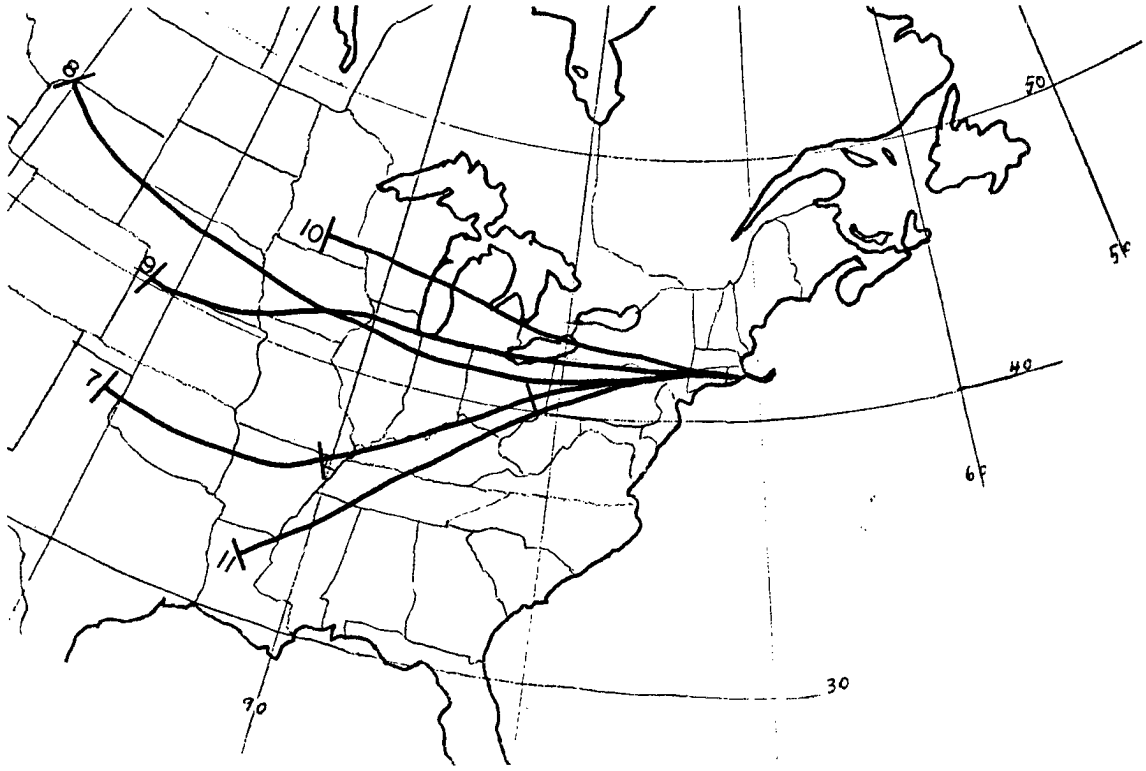


Fig. E.5b Same as a at 700 mb.

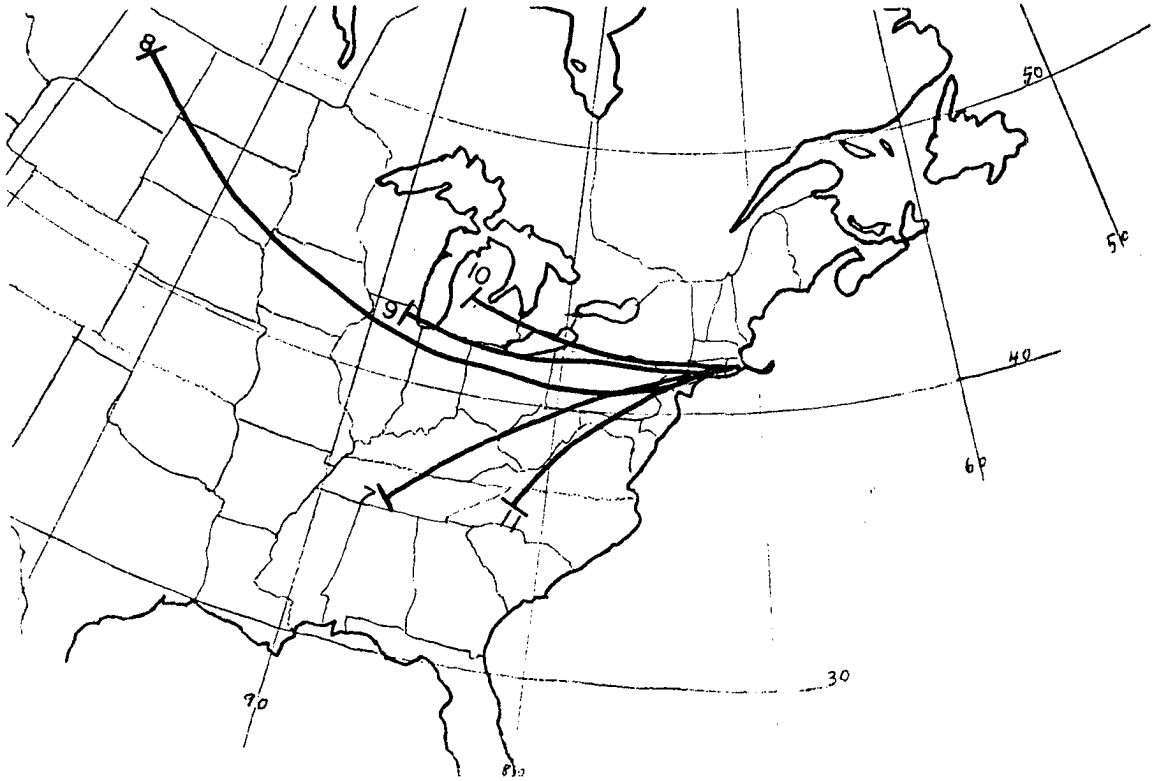


Fig. E.5a Narragansett 850 mb isohypse analyses, 7 - 11 January, 1980, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

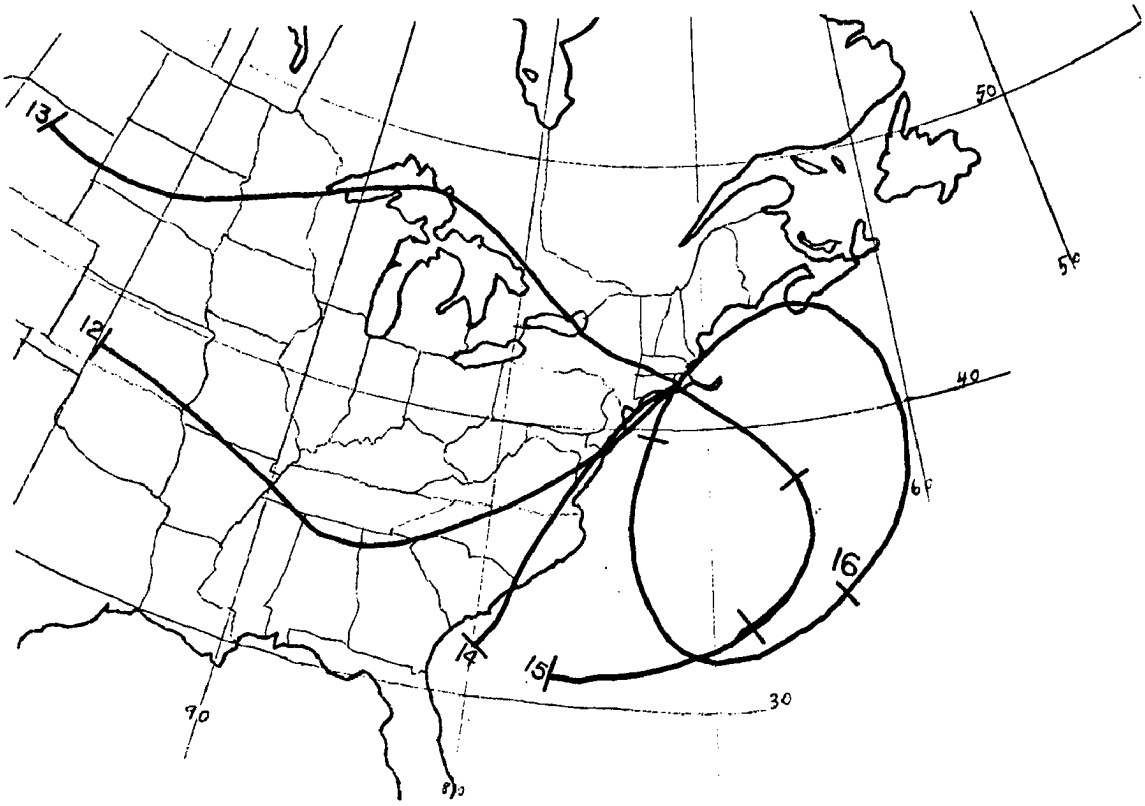


Fig. E.5d Same as a at 700 mb, 12 - 16 January, 1980.

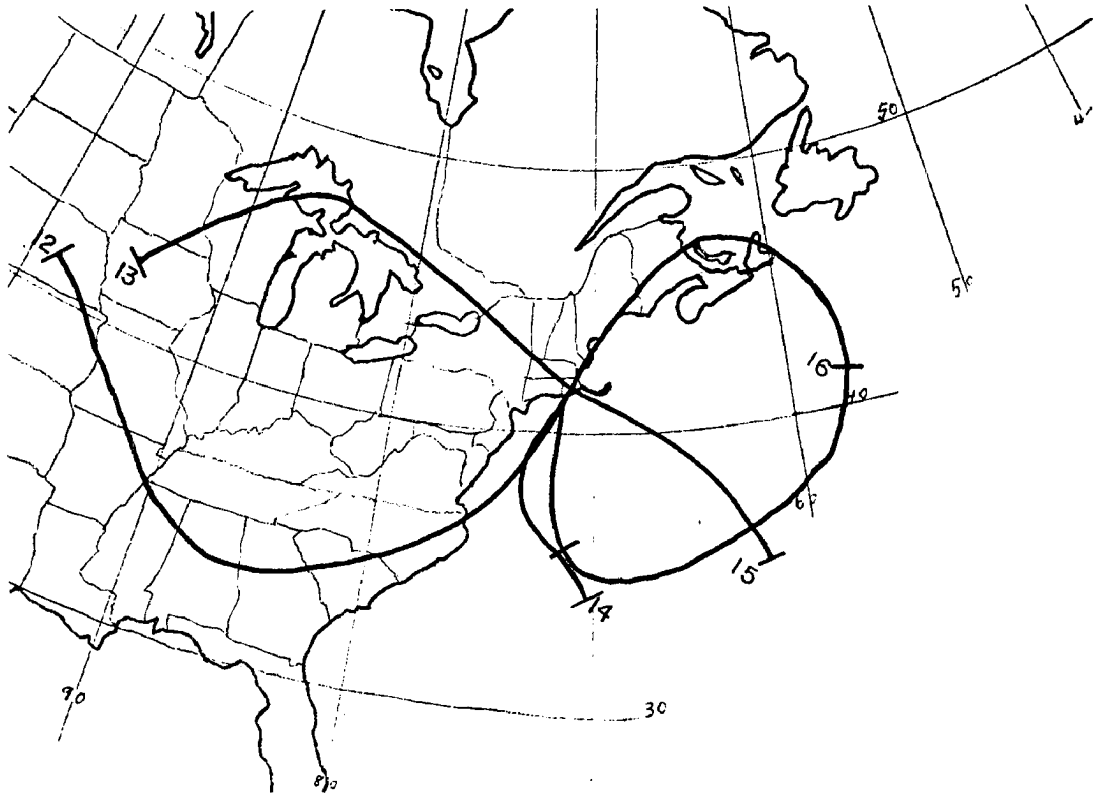


Fig. E.5c Same as a at 850 mb, 12 - 16 January, 1980.

Comparison of Figures E.1 and E.4 shows the effect of the different meteorological conditions of summer and winter on the aerosol trace element chemistry. There is much more variability in the aerosol during the winter. This is a reflection of the higher wind speeds and faster moving storm systems at that time. This is also a stormy season for this site. The pollutant tracers $SO^{=}_4$ and V, although more variable with time, are within the same concentration range from summer to winter. Fresh local pollution sources, as indicated by the concentrations of CN which approached daily means of 80,000 cm^{-3}, were much more prevalent during the winter. This was due to the widespread use of wood stoves at the time for residential space heating. The higher wind speeds associated with coastal storms are reflected in the marine Na concentrations when onshore winds occurred. The concentration of marine Na was the 3 to 8 times higher during the winter study. The higher wind speeds are indicated by the distance between hatch marks on the isohypse analysis in Figure E.5. Also because of higher humidities and more frequent precipitation the natural dust loading as indicated by the Al concentrations was from 2 to 9 times lower during the winter study. The reason the pollutant aerosol indicators essentially remained the same from season to season is the combination of the increase in overall combustion rates as a result of the heating season and a decrease in the depth of the mixing layer. Thus the effect of more vigorous circulation and ventilation offsets the higher emission and lower vertical mixing.

During the period, three separate pulses of marine aerosol particles were advected to the site. One at the beginning, middle and end. These are inversely correlated with the concentration of Al. The

marine source of I is suggested by the parallel variation of I with the marine Na. The non-marine $SO^{=}_4$ does not undergo much variation with time. There is a slight indication that the $SO^{=}_4$ is inversely related to the marine Na. The non-crustal V also shows an inverse relationship to the marine Na during the first two thirds of the study. During the last third of the study, samples 7-10, the V increases along with the marine Na. During this period, an intense low pressure center moved off shore south of the study site. Although wind direction and the isohypse analyses indicate flow from the south through northeast, a maritime direction, the primary pollutant indicator, V, shows that a polluted air mass was advected off shore and returned to the sampling site around the low pressure center. Precipitation was none to light and haze was observed.

The samples for the SEM photographs in Figure E.6 were chosen based upon the high relative pollutant-continental character of sample 3, and the low pollutant-continental character of sample 6. This was based upon the concentrations of Al, V, CN and Na as shown in Figure E.4. Clearly, Figure E.6b shows an oil fly ash particle (Griffin and Goldberg, 1979). The high magnification aerosol photograph, Figure E.6a, shows the filter surface to be entirely coated with a uniform film with other particles superimposed on and within it. Much of this material is suspect of being organic in nature. Figues E.6c and d show the film to be lacking. The broad, flat aerosol particles, presumed to be organic, are present as well as crystalline particles, which are most likely sea salt particles and other salts. In this case, the photographs support the elemental analysis conclusion that sample 3 has a greater mass loading than sample 6.

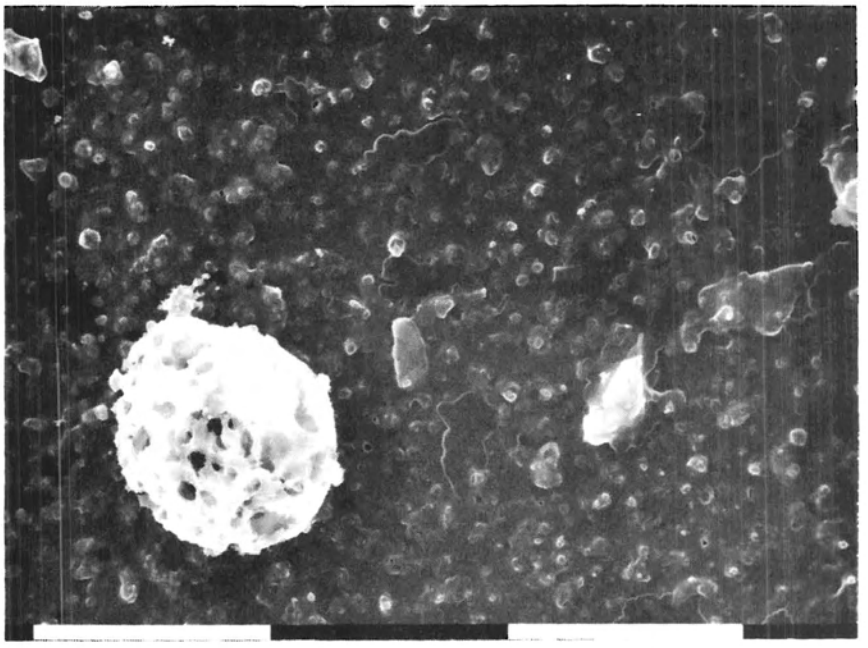


Fig. E.6a Scanning electron microscope photographs of aerosol particles collected at Narragansett during winter on 0.2 μm diameter pore size Nuclepore filters from sample #3, 3100X magnification, bar scale is 10 μm.

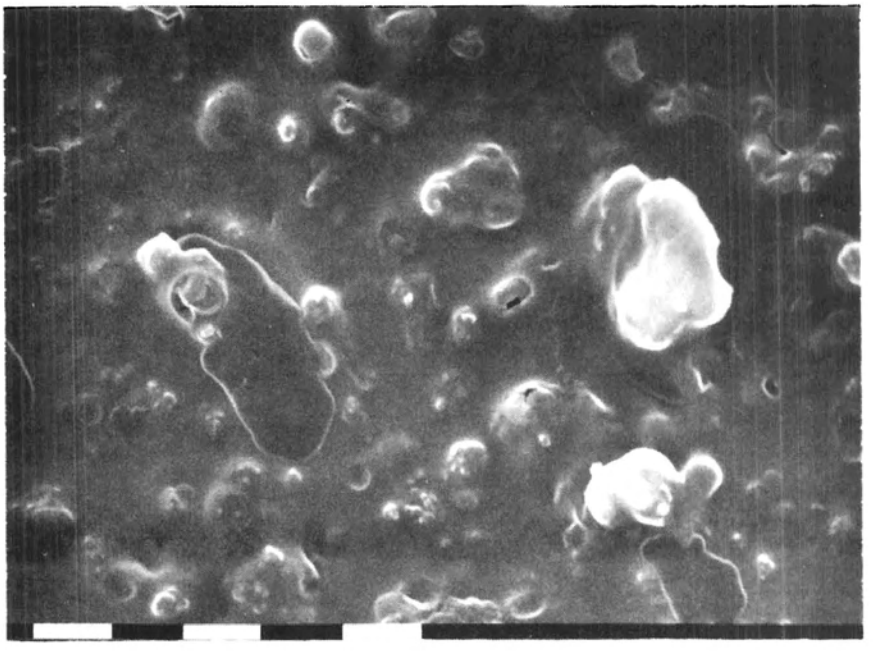


Fig. E.6b Same as a, but 10,000X magnification, bar scale is 1 μm.

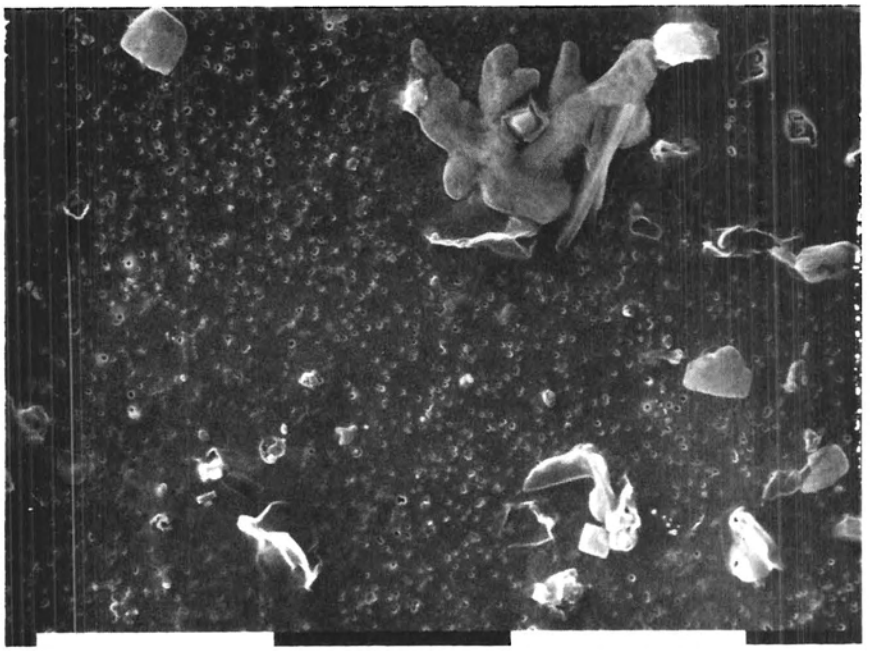


Fig. E.6c Same as a but for sample #6.

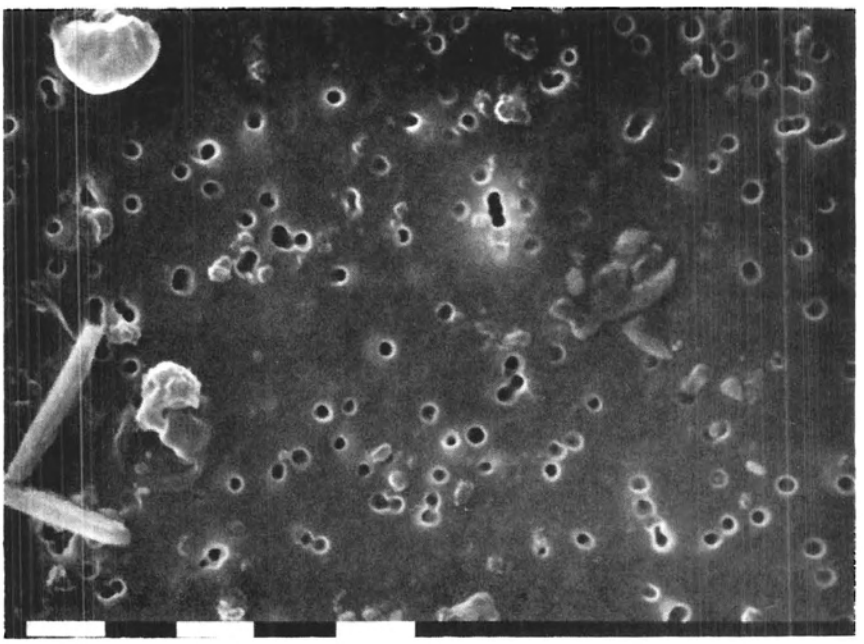


Fig. E.6d Same as c, but 10,000X magnification, bar scale is 1 μm.

E.2a Cloud condensation nuclei

This study period was characterized by more variability of the CCN than the summer case. Table 5.1 shows that most of the CCN variance can be explained by non-marine $SSO^{=}_{4}$ when the entire ten samples are considered. This suggests that the secondary pollutant aerosol particles are a major component of the CCN concentration. This is also supported by looking in more detail at the last four samples for this period. As discussed above, the meteorology and aerosol chemistry indicate that transport of pollutant aerosol particles around an off shore low pressure center occurred between January 14-16. This is when the highest V concentrations and the third peak in the marine Na, occurred, visible haze was reported and the CCN concentrations peaked (sample 9) a second time. This can be considered to be due to unactivated CCN. Based upon this line of reasoning, the CCN concentrations at this site are primarily due to variations in the concentrations of pollutant aerosol as indicated by the primary and secondary pollutant tracers V and $SO^{=}_{4}$.

E.2b Ice nuclei

The analysis of the IN is more straightforward. The per cent variance explained in Table 5.1 for IN in Narragansett during winter show a strong relationship between IN and non-crustal V. The relatively high per cent variance of IN explained by marine Na and I can be interpreted by the following reasoning. The IN are primarily associated with pollutant aerosol particles which were transported over the sea surface. This occurred repeatedly during the study. Each peak in marine Na is associated with the passage of a short wave to the south of the sampling site. In each case, pollutants were transported off shore

on the east to southern sides of the surface low pressure system. Since the sampling site is located 200 km ENE of the major urban-industrial area of New York and New Jersey, it is downwind of the pollution aerosol particle source region. The pollutant transport reaches Narragansett from the marine sector (S to NE) and is therefore associated with a marine background aerosol. The non-crustal V component of the aerosol particle trace element chemistry is linked to emissions from oil fired power plants. Figure E.6b shows an oil fired power plant fly ash particle. Since peak values of IN concentrations are in the range of 0.5 to 1 per liter, the fly ash particles could account for the observed IN variation at this site.

E.3 Seasonal Differences in Cloud Active Aerosol

Figures E.7 and E.8 present the total mean CCN and IN spectra, respectively, for the entire winter and summer study periods. Figures E.8 and E.9 depict the individual sample IN concentrations for summer and winter, respectively, with the mean spectra plotted as solid lines. These spectra are thus representative of the seasonal means for this site. The summer IN spectra reflects the potential problem with the correction factor for the concentrations at $-20^{o}C$. In general, it is clear that the concentrations of both cloud active aerosol particles are higher at all points during the summer than during the winter. Tables 5.2 and 5.3 list the mean seasonal concentration ratios of CCN at 0.2% SS, IN at $-20^{o}C$ and 1% water SS and the aerosol particle pollutant tracers non-crustal V and non-marine Na. Comparison of the data for Narragansett in these tables with Figures E.7 and E.8 reveals that although summer concentrations of CCN and IN are greater than winter concentrations, winter concentrations of the pollutant aerosol are the

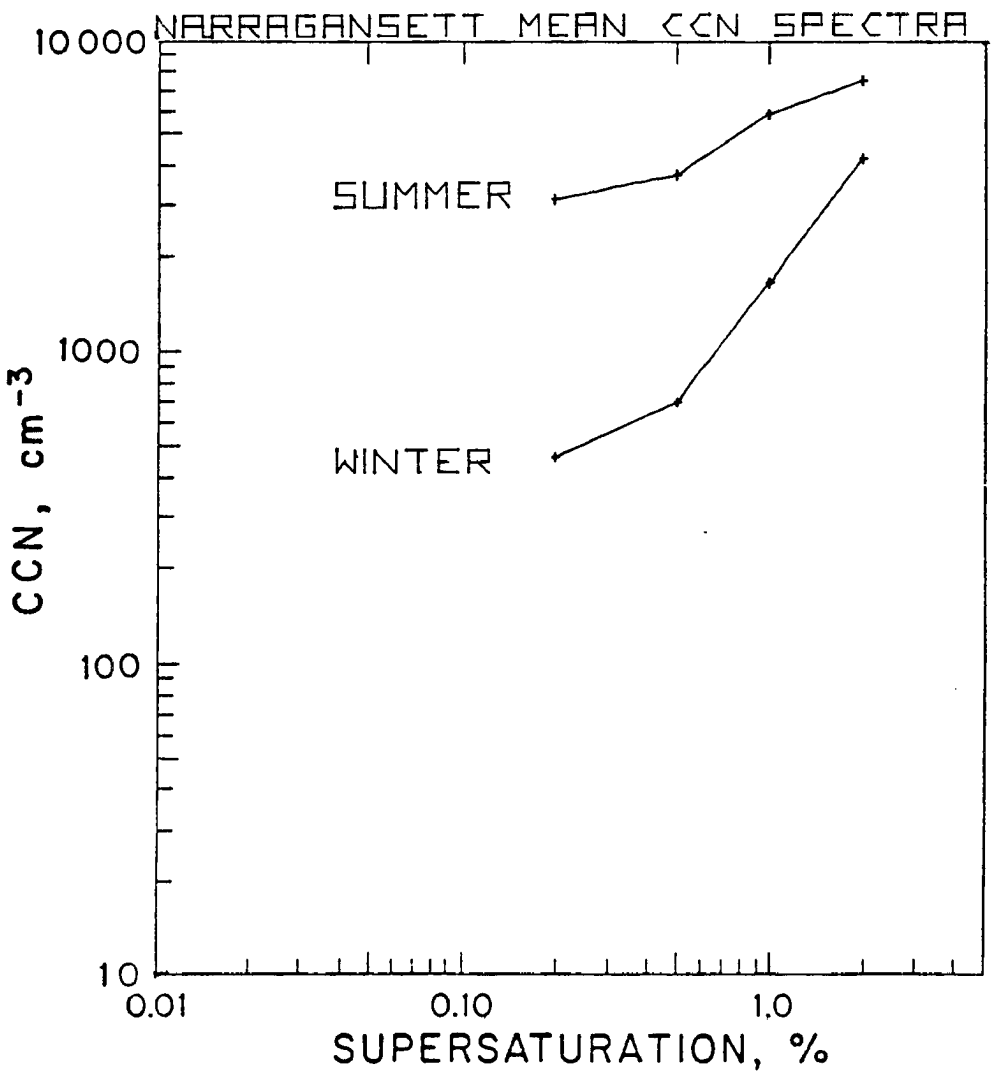


Fig. E.7 Mean CCN spectra for Narragansett, Rhode Island, USA, derived from thirteen spectra measured during summer, 12 -19 June, 1979 and twenty spectra measured during winter, 6 - 15 January, 1980.

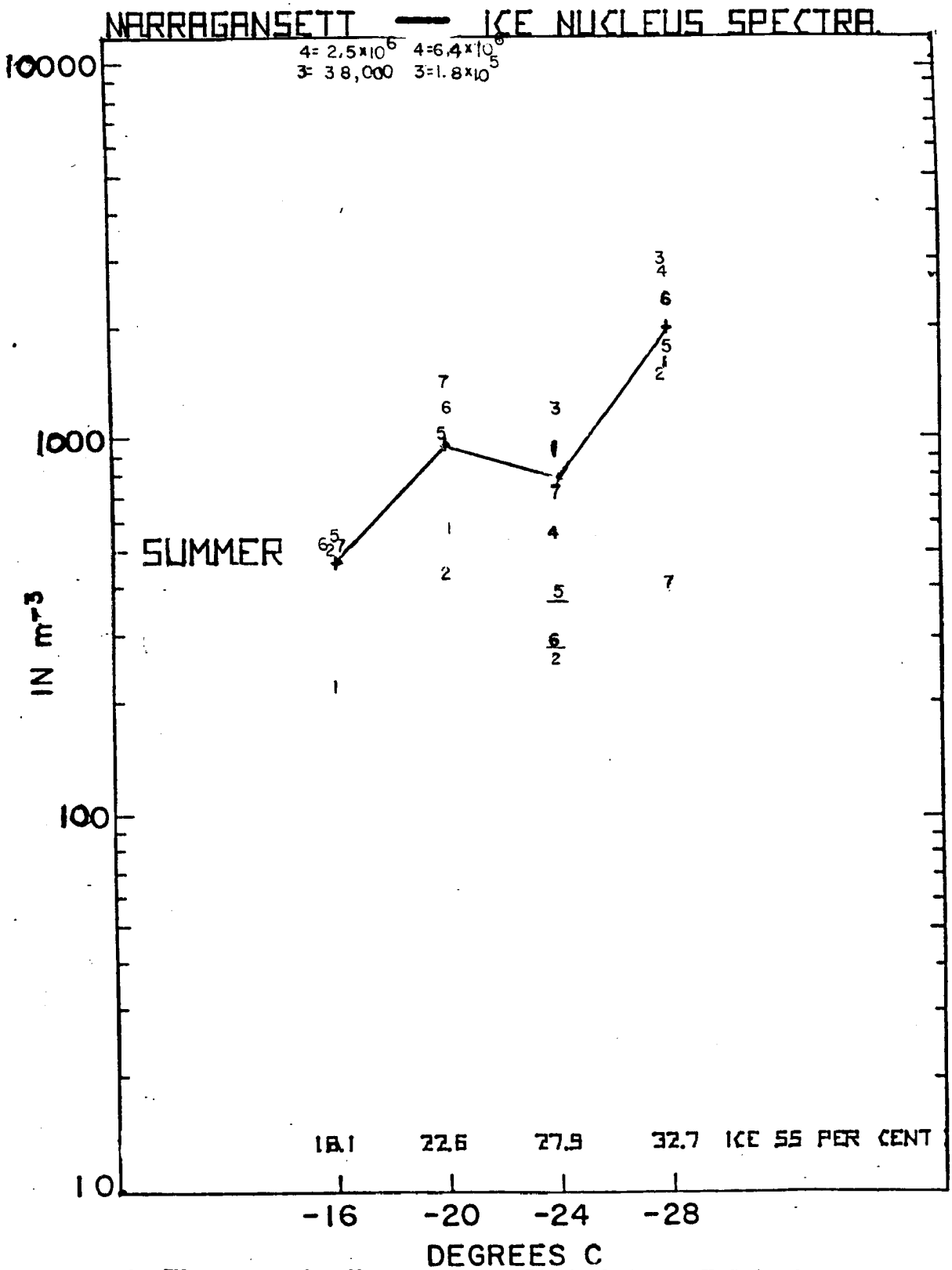


Fig. E.8 IN spectra for Narragansett, Rhode Island, U.S.A. for the summer experiment, 12-19 June 1979. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank, without samples 3 and 4 at teperature $-10^{\circ}C$ and $-24^{\circ}C$.

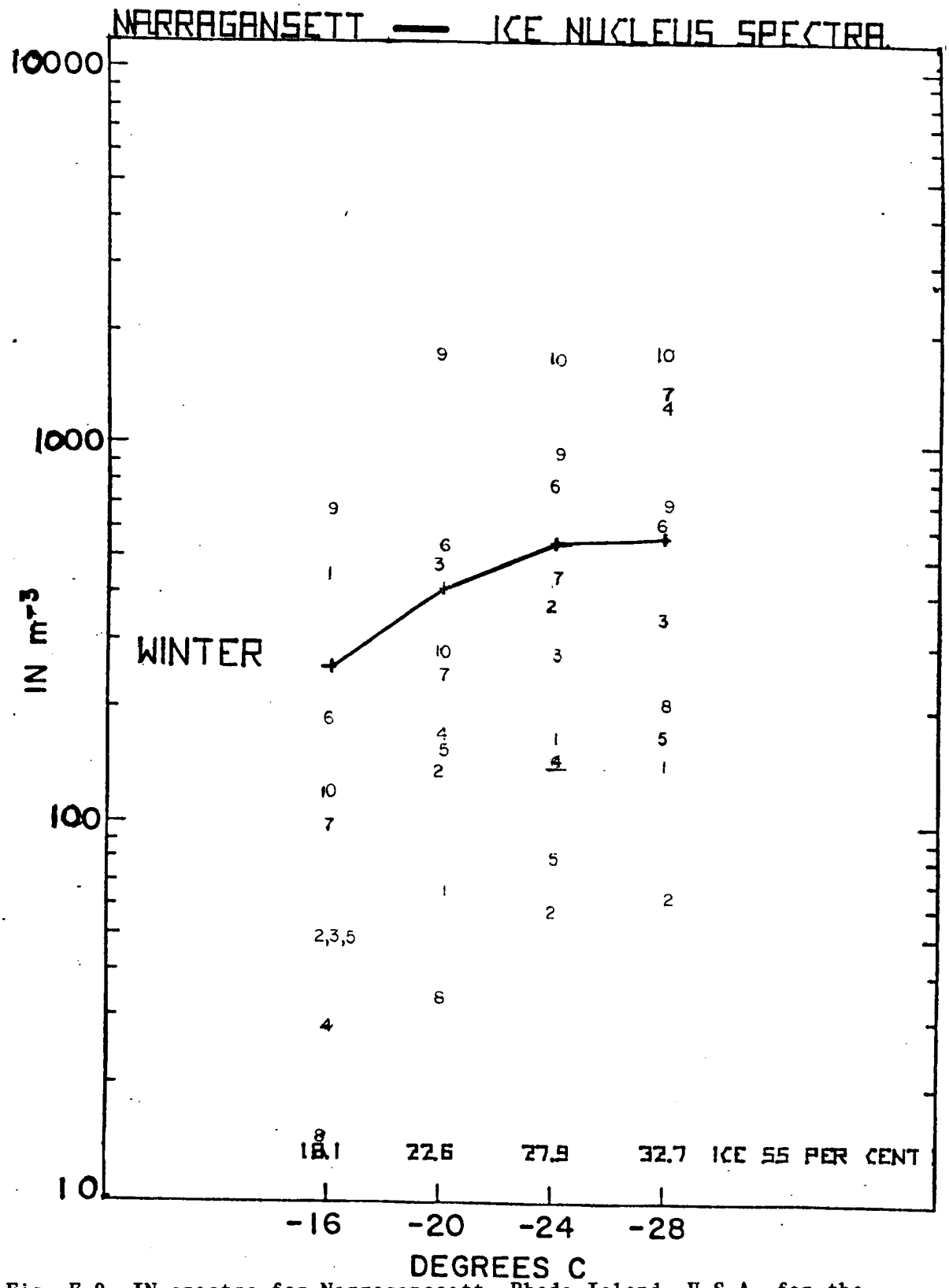


Fig. E.9 IN spectra for Narragansett, Rhode Island, U.S.A. for the winter experiment, 6-15 January 1980. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

same or just slightly greater than those during the summer. Within a given sseasonal sample set, pollutant aerosol particles are the major component of the total cloud active aerosol population. The apparent seasonal discrepancy to this conclusion is due to the different meteorological conditions that exist during winter and summer. As shown in the discussion above, the winter at this location was characterized by stormy conditions. Within a given aerosol population, the cloud active aerosol will be preferentially removed during the formation of clouds and precipitation. This is an importanct point. The basic premise of the aerosol trace element method of tracing cloud active aerosol sources is that the element of interest is not necessarily an actual component of the cloud active aerosol composition. Rather, it is associated with the same aerosol population which has been advected to the study site.

The CCN show a greater seasonal difference than that of the IN. IN, by their nature, are primary aerosol particles, in general, they must be insoluble. CCN are hygroscopic aerosol particles. Thus, the secondary particles formed from the oxidation of SO_2 are important as CCN. This was shown in Table 5.1. Thus, any enhancement of the oxidation of SO_2, will also act to form higher numbers of CCN. Summer clearly has conditions which are more suitable for this transformation process: low wind speeds, higher absolute humidities and strongerr sunlight for longer lengths of time. These factors are certainly a component in the explanation of more than 5 times higher CCN concentrations during summer in comparison to those of winter at this site. The higher summer concentrations of CCN and IN are due to differences in removal processes. With a given reason, the CCN and IN

are primarily a result of regional primary and secondary pollutant emissions.

E.4 Cloud Active Aerosol Sources

From the simultaneous observation of cloud active aerosol particles and aerosol chemistry in conjunction with simple meteorological analysis, several conclusions on the sources of IN and CCN at Narragansett can be drawn. 1) Regional pollution sources can explain most of the variance of the IN and CCN concentrations. 2) The transport path of pollutants to Narragansett has an over ocean portion both during winter and summer. Marine sources of CCN and IN are not important since the pollutant aerosol component still overshadows the marine component along these transport paths. 3) Continental airmasses contain the lowest concentrations of IN and CCN for this location. Based upon these observations, anthropogenic sources of cloud active aerosol dominate the natural sources at this mid-latitude source region site.

APPENDIX F

DETAILED FIELD EXPERIMENT DISCUSSION
Barrow, Alaska, USA

This site was chosen by the National Oceanic and Atmospheric Administration (NOAA) as one of the four baseline monitoring stations for the Global Monitoring for Climatic Change (GMCC) program. It is an excellent aerosol experiment station. The site is located with the Arctic Ocean on the west, north and east sides within 3-4 km. During the winter, snow and ice cover both the land and sea surface. During summer, there is typically more than 100 km of open water between the coast and the ice offshore. Essentially uninhabited tundra extends 250 km south to the Brooks Range, separating the North Slope of Alaska from the interior. The village of Barrow, population 2500, lies 7 km to the southwest of the sampling site. The prevailing wind direction is northeast. The Prudhoe Bay-Deadhorse complex lies 280 km to the southeast.

F.1 Summer Study

The summer study period extended from 28 July to 7 August, 1979. Ten 24-hour sample sets were collected and analyzed. The results of the bulk-aerosol trace-element composition are given in Table 3, Appendix A. Figure F.1 is a plot of the day-to-day variation of the elemental source tracers and the CCN and IN concentrations at 0.2% SS and -20°C, 1% SS respectively. Figures F.2a-d are isohypse analyses for the study period for the 850 mb and 700 mb pressure surfaces. The same notation scheme

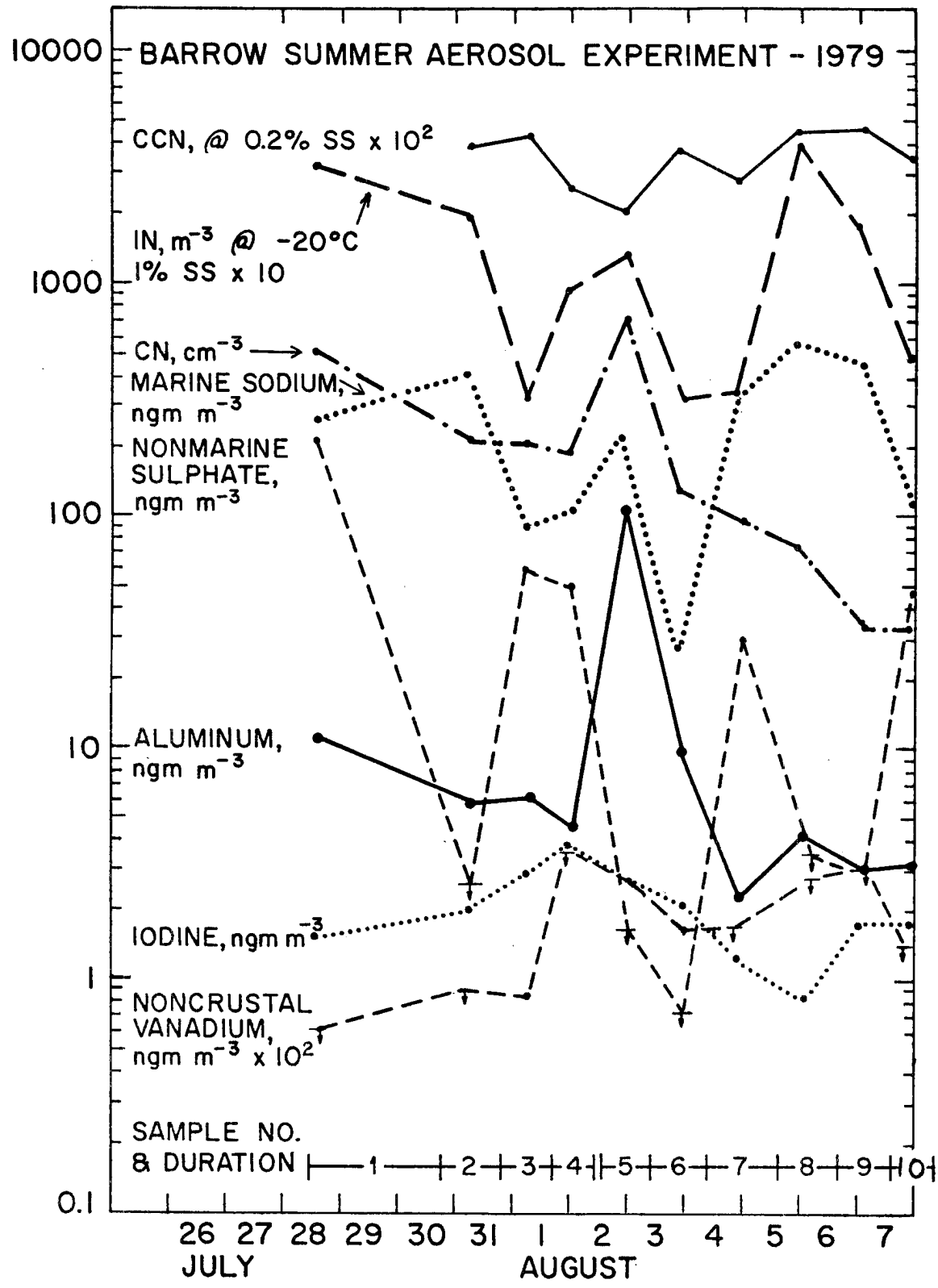


Fig. F.1 Barrow, Alaska, USA summer aerosol experiment, July - August 1979.

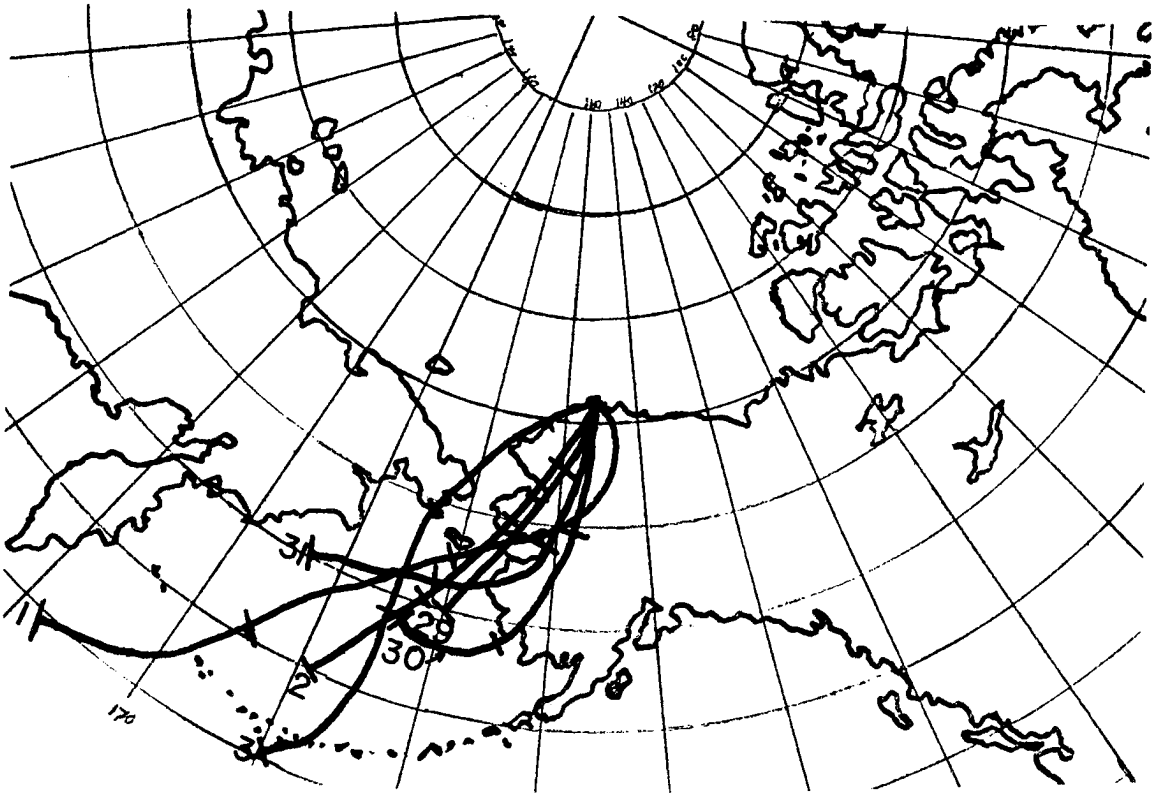


Fig. F.2b Same as a at 700 mb.

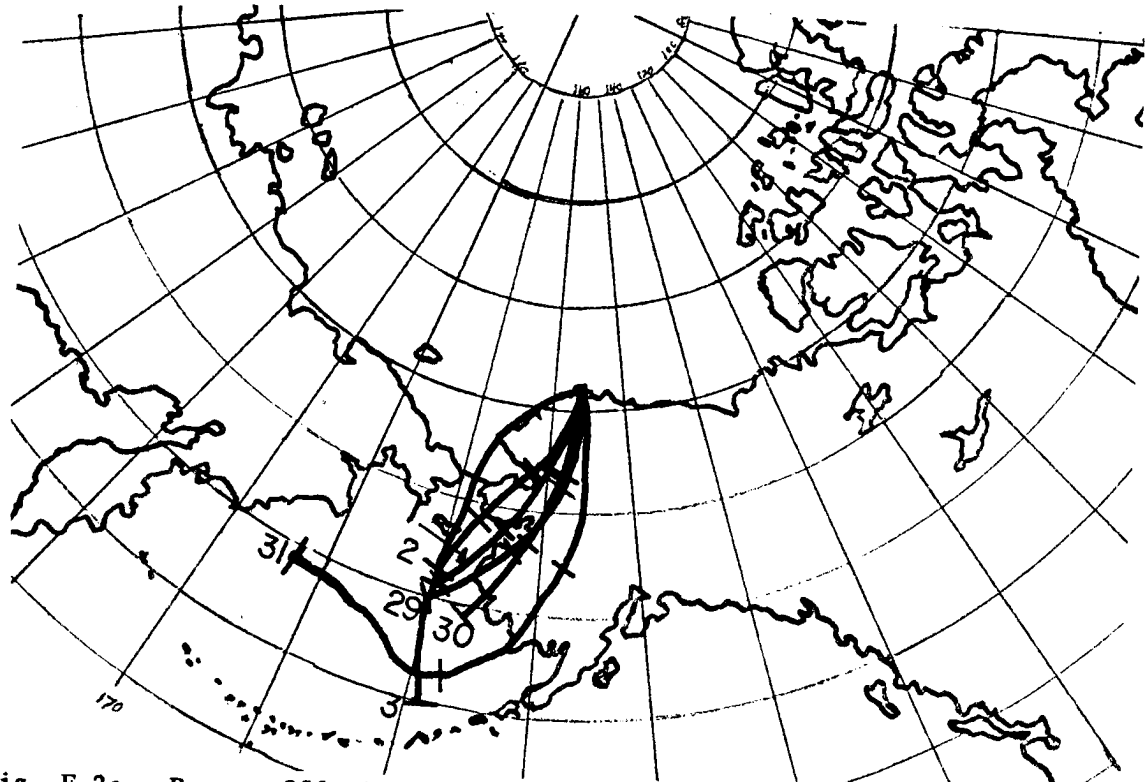


Fig. F.2a Barrow 850 mb isohypse analyses, 29 July - 3 August, 1979, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

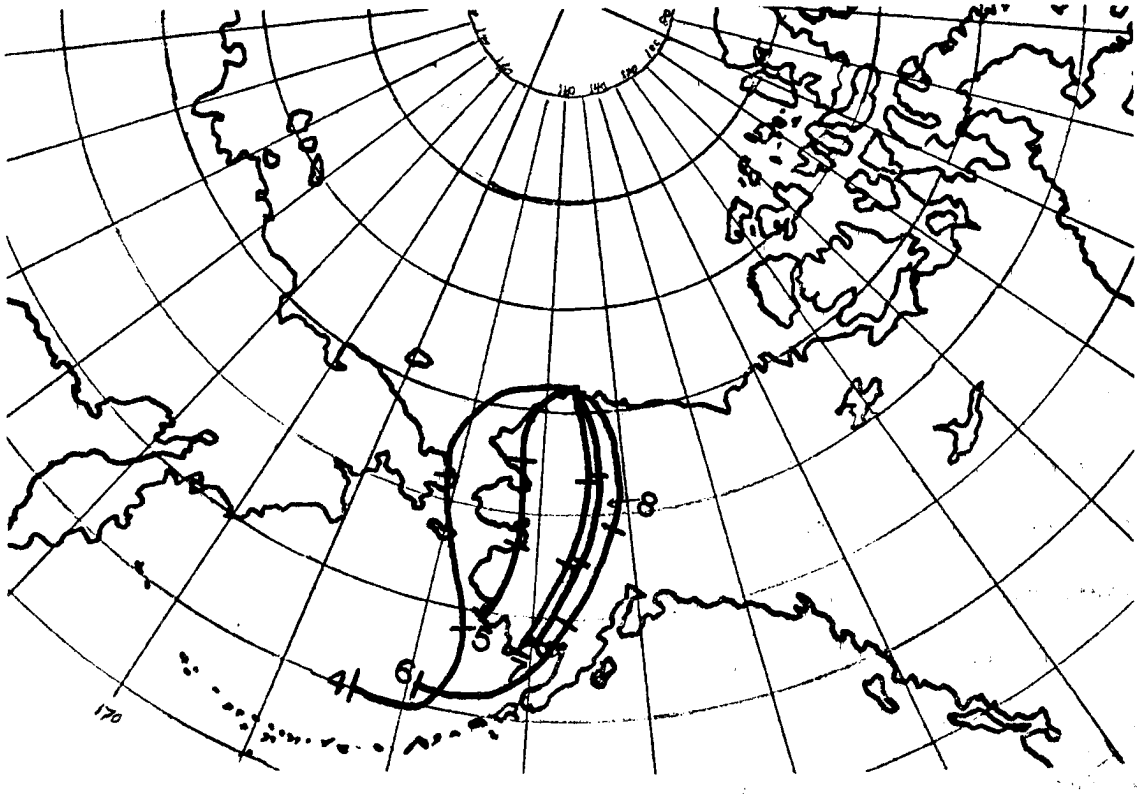


Fig. F.2d Same as a at 700 mb, 4 - 8 August, 1979.

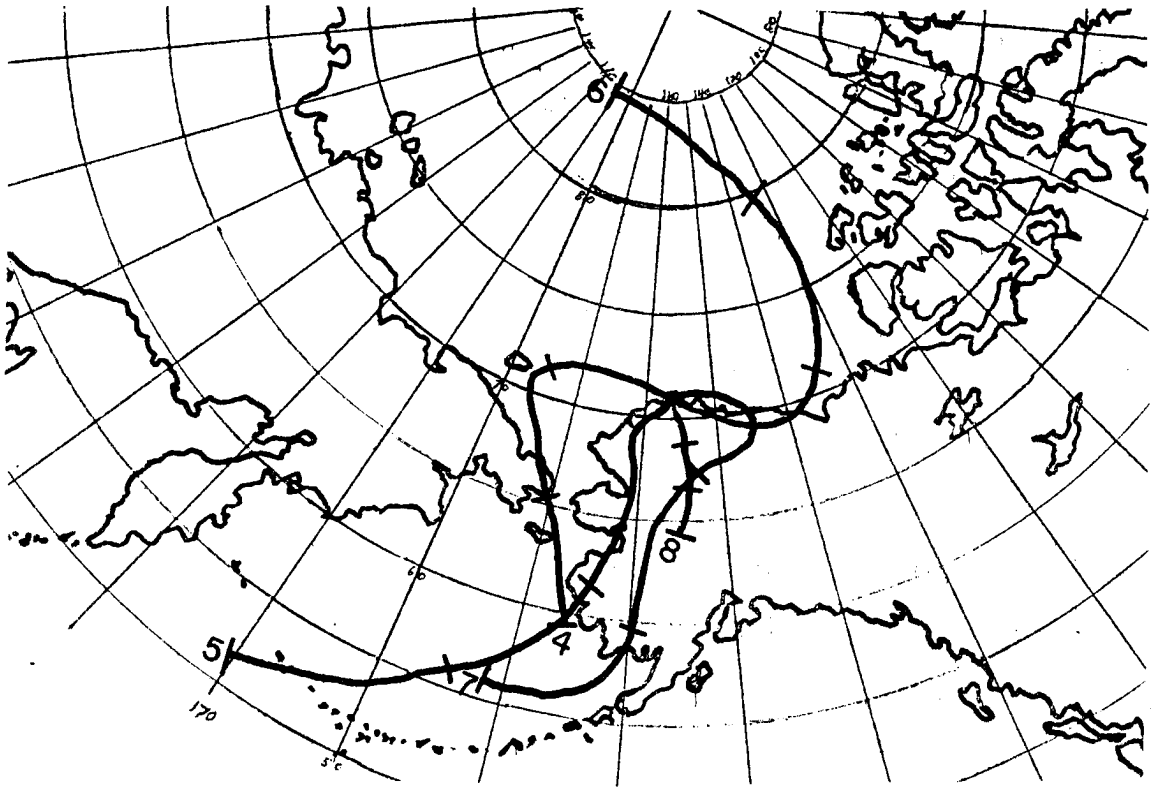


Fig. F.2c Same as a at 850 mb, 4 - 8 August, 1979.

used for the Narragansett study in Appendix E aplies to these isohypse analyses.

The aerosol particle concentration is low at this remote background site. The pollutant aerosol particle indicators V and $SO_4^=$, when detected, were 10-100 times less in concentration in comparison to the measurements at Narragansett. Because the site is located on the Arctic Ocean coast, the marine aerosol indicators, I and marine Na, are less than a factor of 10 lower in concentration in comparison to the mid-latitude site. CN concentrations, reflecting fresh local combustion sources, are as low as 40 cm^{-3} compared to a low of 3000 cm^{-3} at Narragansett. These differences between the aerosol particle populations of the two sites are reflected in the SEM photomicrographs for samples 1 and 9 shown in Figures F.3a-d at magnifications of 3100X and 10,000X. The volume of air sampled for sample 1 in these photographs is 2.5 times higher than that of sample 9. This explains part of the greater numbers of particles on the sample 1 filter.

The winds at Barrow below the surface temperature inversion were typically northwest to northeast during the period. The isohypse analysis shows the winds aloft were almost entirely southwesterly. Sample 5 was a case of local contamination as evidenced by the peak in CN concentrations. This will be discussed further in following sections.

F.1a Cloud condensation nuclei

The CCN concentrations were observed to be 10 to 100 times lower in concentration at Barrow than at the mid-latitude source region site, Narragansett. Although many of the aerosol particle tracers depicted in Figure F.1 underwent a great deal of variation with time, the CCN

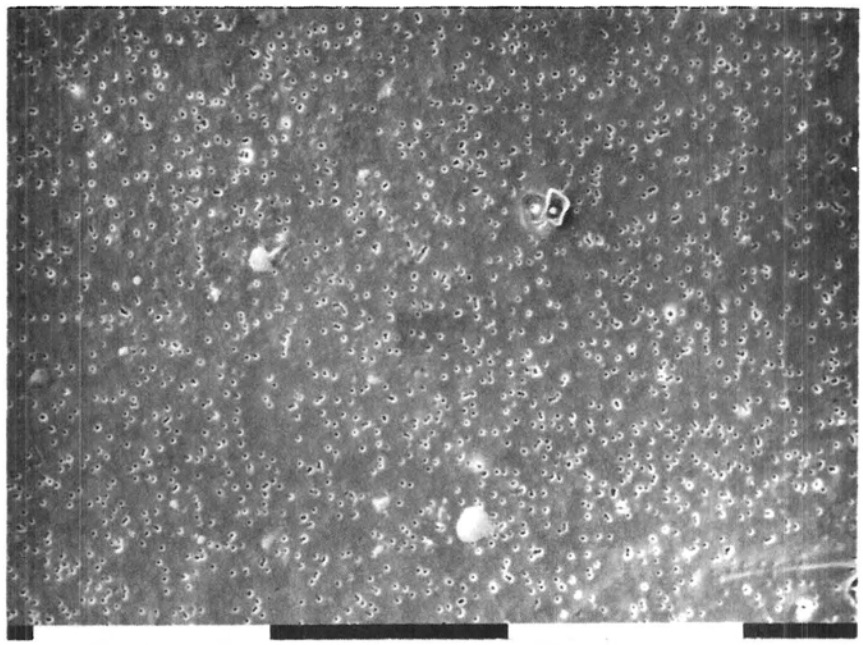


Fig. F.3a Scanning electron microscope photographs of aerosol particles collected at Barrow during summer on 0.2 μm diameter pore size Nuclepore filters from sample #1, 3100X magnification, bar scale is 10 μm.

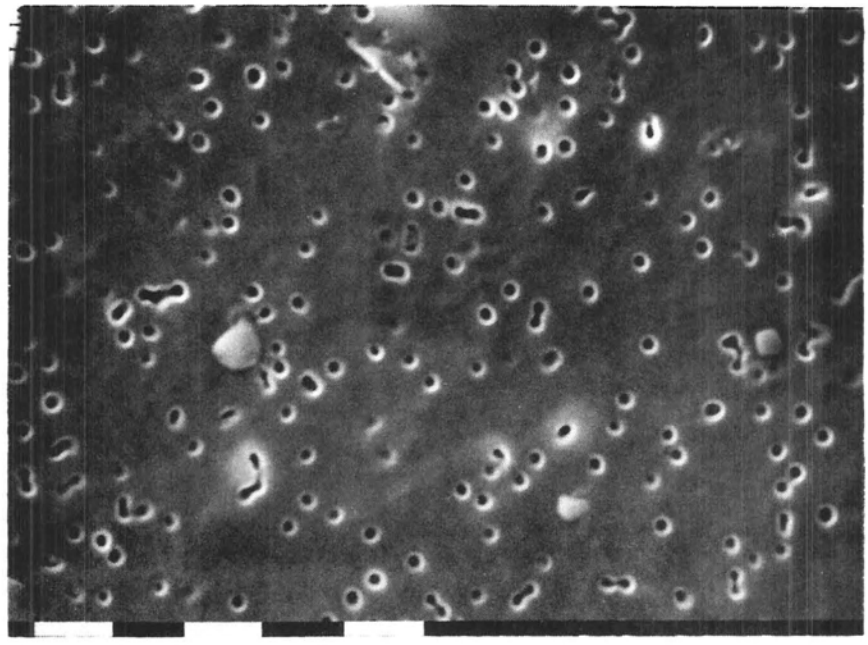


Fig. F.3b Same as a, but 10,000X magnification, bar scale is 1 μm.

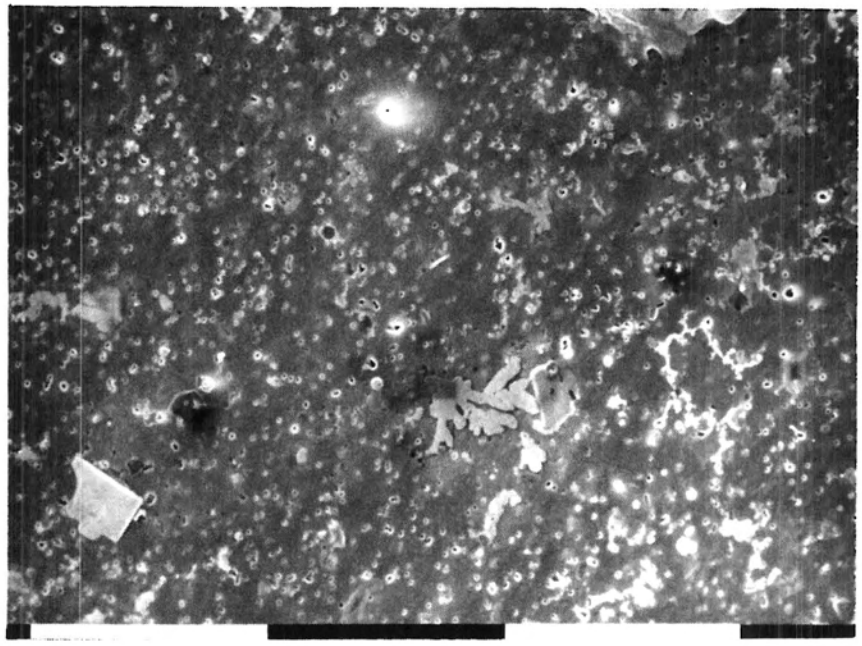


Fig. F.3c Same as a but for sample #9.

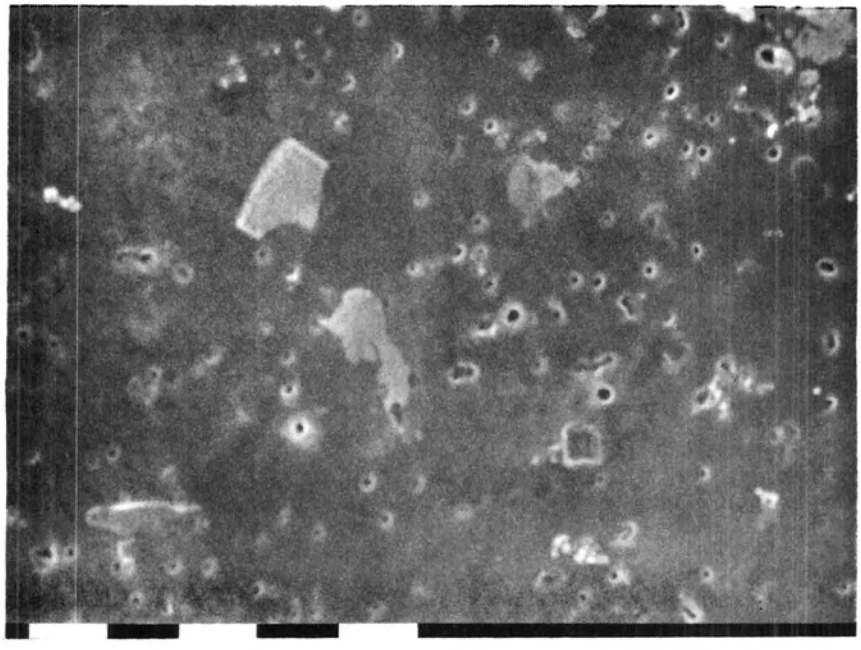


Fig. F.3d Same as c, but 10,000X magnification, bar scale is 1 μm.

concentrations were fairly stable between values of 20-45 cm^{-3} at 0.2% SS. Since the pollution aerosol elemental tracers were not detectable the majority of the time, no definitive statement of the relatonship between the CCN and these aerosols can be made. Referring to Table 5.2, the variance of the CCN is not significantly explained by any of the elemental tracers. There is an indication that primary marine aerosol may be a component of the CCN. The only positive correlation occurs between CCN and marine Na. Other components of the CCN concentrations can only be speculated upon. Figure F.3a and b show that the same-assumed organic aerosol particles as seen in the photomicrographs from Narragansett, exist at Barrow or were an artifact of sampling. These may be a component of the CCN concentrations. The secondary-marine aerosol particle tracer, I, does not provide any indication of a relationship with the CCN. If organic species are important, these data are not sufficient to draw any conclusions.

Sample 5, the locally contaminated sample, provides useful information on the effects of local anthropogenic aerosol particles on the CCN concentration. For this sample, the CCN are at their lowest and the CN at their highest concentration for the period of study. This is strong evidence that local sources of fresh aerosol particles do not increase the CCN concentrations, but in fact may cause a decrease. The results in Table 5.2 support this conclusion. The correlation coefficient between CCN and CN was -0.62, significant at the greater than 90% level, the most significant negative correlation within the data set.

F.1b Ice nuclei

The IN concentrations were more variable than the CCN concentrations suggesting the influence of different sources, or the variation of a single source. The overall concentration of IN at $-20^{o}C$ is a factor of 10 to 10^{4} less than that measured at Narragansett depending on whether the values for samples 3 and 4 from Narragansett are used or not. Table 5.2 shows that 56% of the variance of the IN at $-20^{o}C$ can be explained by the primary marine-Na tracer. All the other tracers show little to no relationship to the IN concentrations. This suggests a marine source for IN near Barrow during the summer. Pollutants transported over long-range, or from local sources, either do not exist or are not important in explaining the variation of the IN at this site.

The possibility of marine sources of IN has been suggested by other investigators. The most relevant study was reported by Jayaweera and Flanagan (1981). Their results from aircraft measurements of IN over the Arctic Ocean near Barrow during summer showed the presence of biogenic IN active at temperatures as warm as $-6^{o}C$ and at concentrations of 1 m^{-3}. This is in line with the results presented here since they also reported the concentrations decreased near the surface and their reported concentrations were at a temperature $10^{o}C$ warmer than the coldest temperature studied in this experiment. Concentrations of IN at $-16^{o}C$ from this study had an average value of 78 m^{-3}. Results from the isohypse analyses suggest the Arctic Ocean may be the most important source of IN. Most of the time the flow aloft was from the southwest. For one day, the winds up through the 850 mb level were easterly and had a history which was over the Arctic Ocean. Sample 8 corresponds to that

day. Note that the marine Na and the IN concentrations have their highest values on this day. Open water and moderate winds were present at the time. The biogenic nature of the IN collected at this site can only be speculated upon. Evidence suggests a marine source.

F.2 Winter study

The Barrow winter study period extended from 24 March to 3 April, 1980. Ten, 24-hour samples sets were collected and analyzed. The results of the bulk- aerosol trace-element composition are given in Table 4, Appendix A. Figure F.4 is a plot of the day-to-day variation of the elemental source tracers and the CCN and IN concentrations at 0.2% SS and -20°C, 1% WSS respectively. Figures F.5a-d are isohypse analyses for the 850 mb and 700 mb pressure surfaces for the study period. The same notation scheme applies to these analyses as described previously.

Barrow is well within the arctic polar airmass during the winter. Although samples were collected during the early spring, this period of the year has been shown to be the time of the annual maximum pollutant aerosol concentration (Rahn and McCaffery, 1980). The difference in the aerosol climate between summer and winter at this site is apparent from comparison of Figures F.1 and F.4. Note that the pollutant aerosol tracers V and $SO_4^{=}$ have average concentrations >45 and >38 times higher, respectively, during the winter than during the summer. Concentrations of Al and I are within a factor of two of the summer values. Marine Na is about the same during the first half of the study period and goes through a maximum on sample 9 which is six times greater than any concentration measured during the summer experiment.

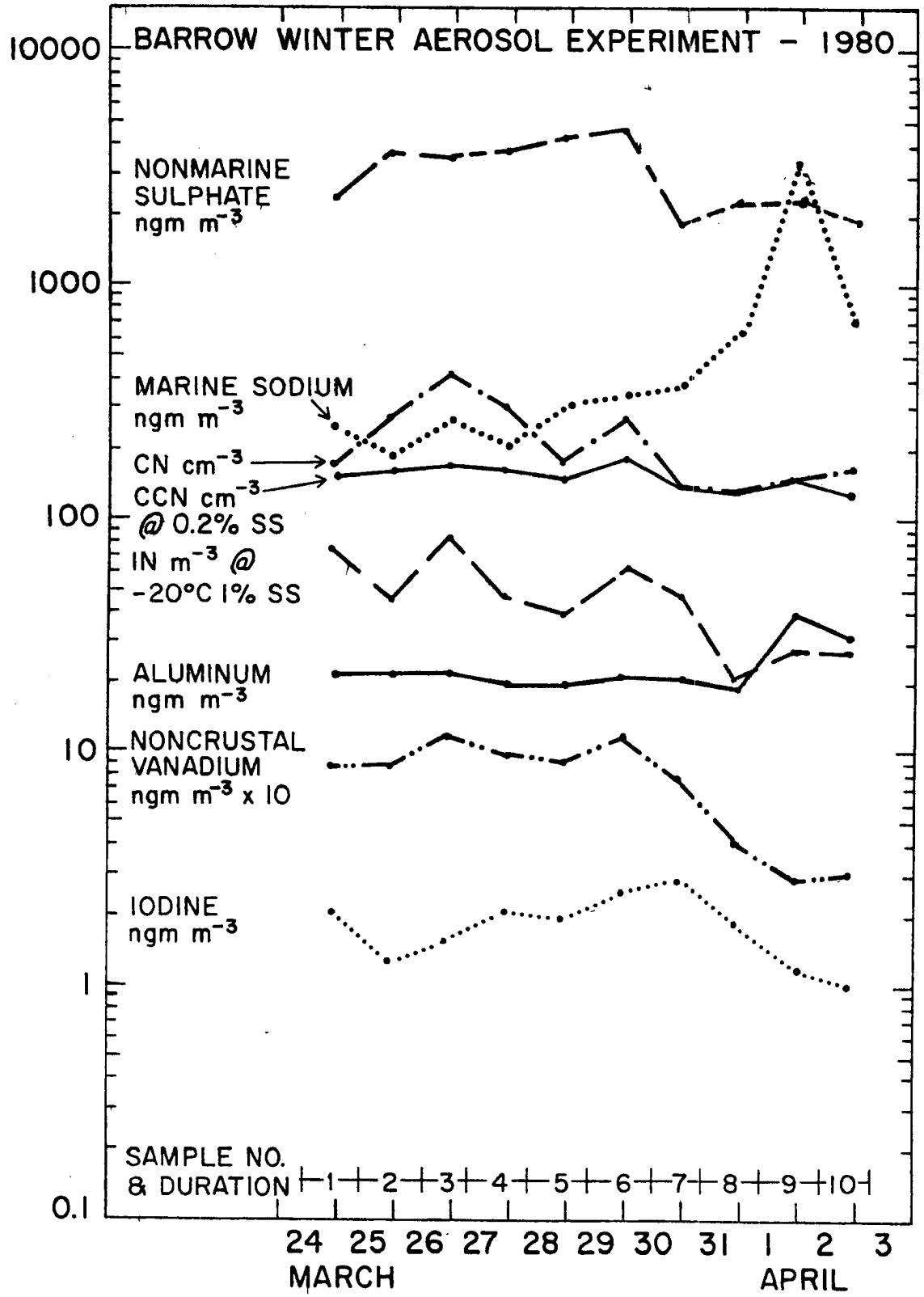


Fig. F.4 Barrow, Alaska, USA winter aerosol experiment, March - April 1980.

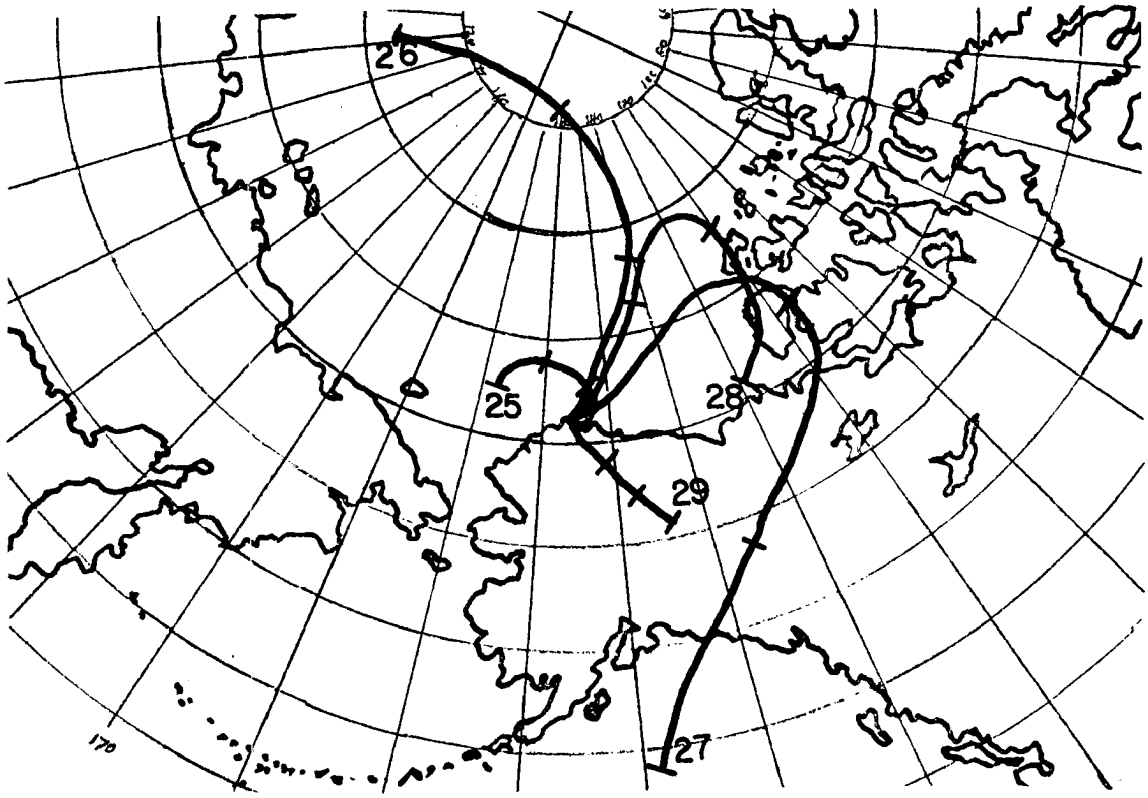


Fig. F.5b Same as a at 700 mb.

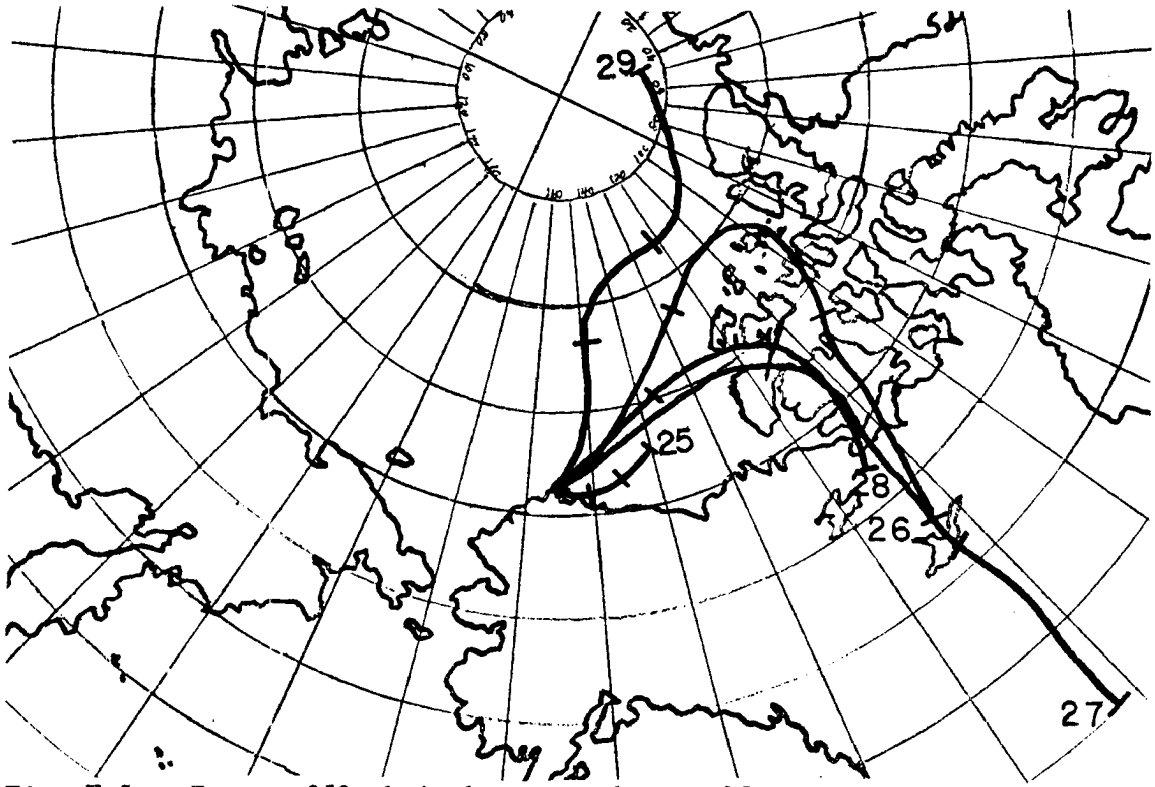


Fig. F.5a Barrow 850 mb isohypse analyses, 25 - 29 March, 1980, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

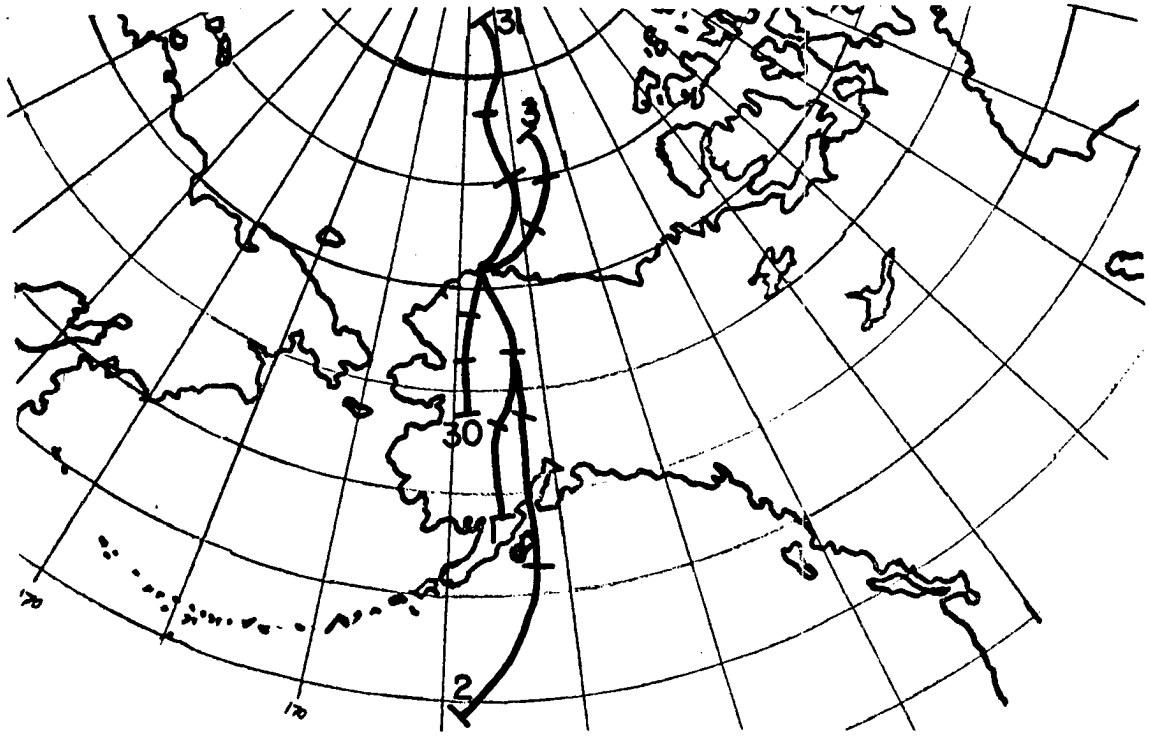


Fig. F.5d Same as a at 700 mb, 30 March - 3 April, 1980.

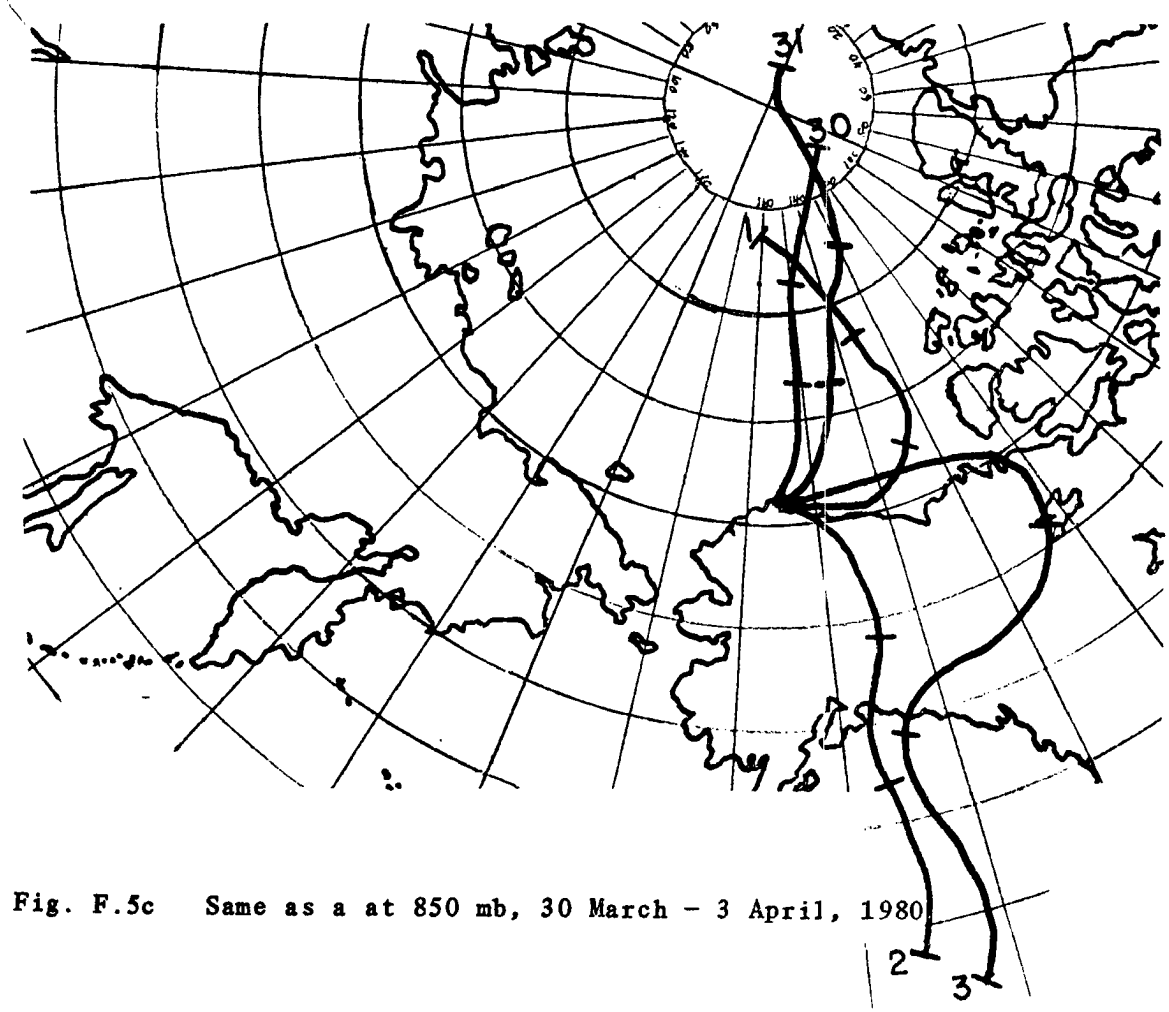


Fig. F.5c Same as a at 850 mb, 30 March - 3 April, 1980.

All of these variations in the aerosol particle composition are due to the meteorological situation. Surface conditions were relatively stable. Winds were consistently between 040° and 090° with speeds 20-30 kts. Surface temperatures ranged from -24°C on 24 March to -32°C on 30 March with a slight warming to -26°C by 3 April. This is reflected in the isohypse analyses in Figure F.5. Flow from the south aloft gradually backed to the north through east to south again by the end of the period. Above the surface temperature inversion, which averaged 1000 m deep with a 10°C temperature difference, the temperature followed the same temporal variation as at the surface. At 850 mb, the temperature ranged from -11°C to -19°C but with more rapid response as the wind direction aloft changed. The surface temperature inversion has a dampening effect on the changes observed at the surface which result from changes of airmass aloft due to the synoptic situation.

The synoptic pattern was dominated bythe Aleutian Low in the North Pacific to the south and west of Barrow and a high pressure system located to the north and west of Barrow. As the Aleutian Low would build in intensity and move toward the northeast, it would displace the high pressure center toward the northwest. The Aleutian Low began moving north beginning around 1 April. Its influence was just waning at the start of the sampling period on 24 March as a result of a previous advance northward. The high pressure center dominated the synoptic flow during the intervening period, and was responsible for the low temperatures on 30 March.

The effect of the changing synoptic situation can be seen in the change in the concentration of the pollutant aerosol tracers, V and $SO_4^=$. During the middle of the study period, both of these tracers have

concentrations that are about 2-3 times higher than during the last four samples collected. This shows the gradual dilution of the cold, polluted, arctic- airmass from the north by the North Pacific airmass transported from the south, a clean "background" airmass in comparison. The maximum in marine Na in sample 9 is not due to sea salt particles transported over Alaska to Barrow. During the collection of this sample, blizzard conditions existed. Winds exceeded 35 kts from the northeast at the surface with blowing and drifting snow. This maximum in marine Na can best be explained by the ablation of sea ice upwind of the sampling site by wind-blown snow and ice crystals. The downward mixing of airmasses above the surface temperature inversion is detectable by changes in the aerosol composition even though surface winds are invariant.

It is interesting to note that Al is also elevated in sample 9 whereas non-crustal V is not. This suggests there may be a fractionation process acting upon these elements during the wet and dry deposition of airborne materials at this site. This phenomenon, a preferential deposition of Al compared to V, has been suggested and discussed by Rahn and McCaffery (1979).

Figures F.6a-d are SEM photomicrographs of the aerosol particles collected during samples 1 and 10. Sample 1 shows the higher aerosol loading associated with the arctic airmass just being advected to Barrow at this time. The aerosol loading on sample 10 is much less and represents the Northern Pacific airmass that had arrived in Barrow 2-3 days earlier. Aerosols from sample 1 were investigated by x-ray analysis using the electron microprobe on the SEM. The analysis revealed the aerosol particles which appear gray or semi-transparent in

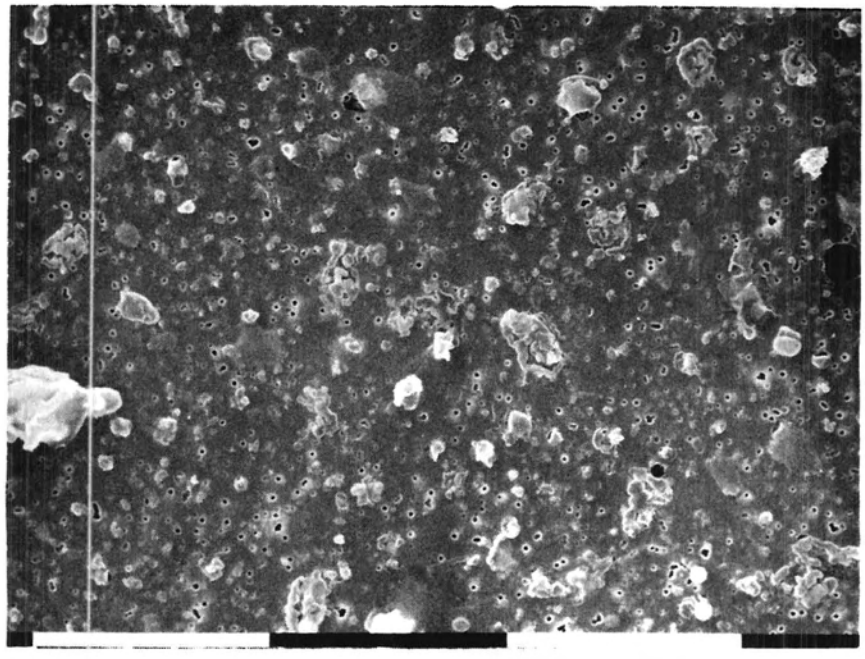


Fig. F.6a Scanning electron microscope photographs of aerosol particles collected at Barrow during winter on 0.2 μm diameter pore size Nuclepore filters from sample #1, 3100X magnification, bar scale is 10 μm.

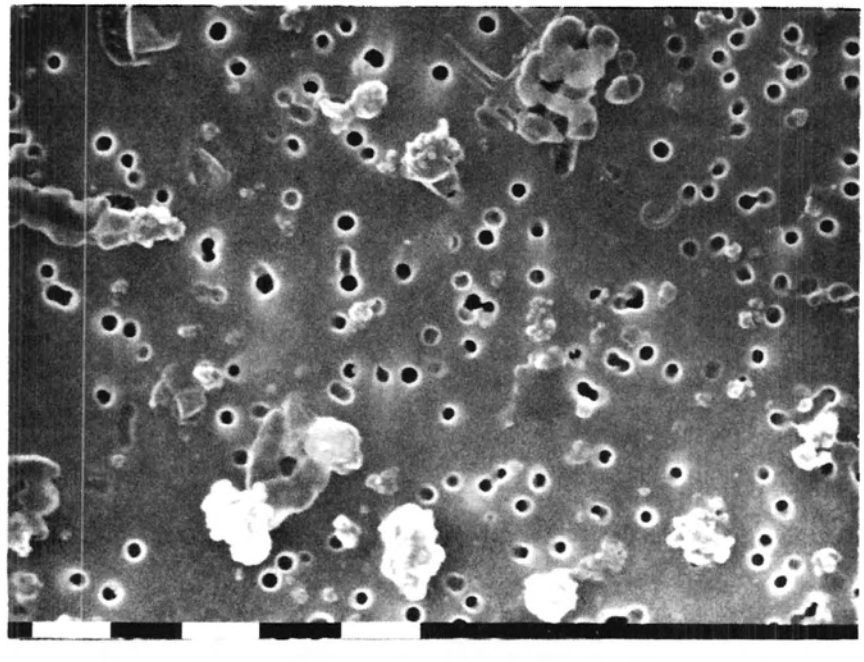


Fig. F.6b Same as a, but 10,000X magnification, bar scale is 1 μm.

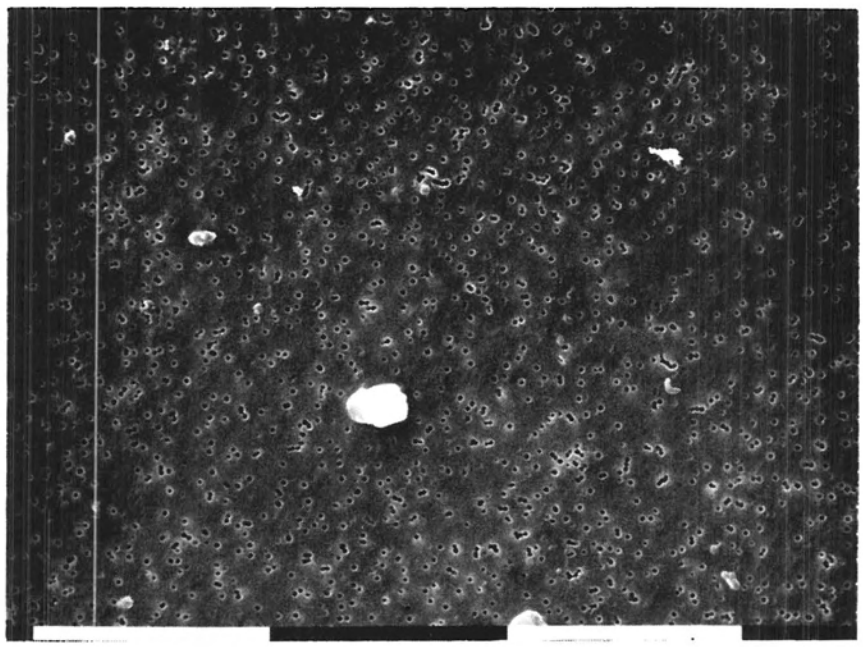


Fig. F.6c Same as a but for sample #10.

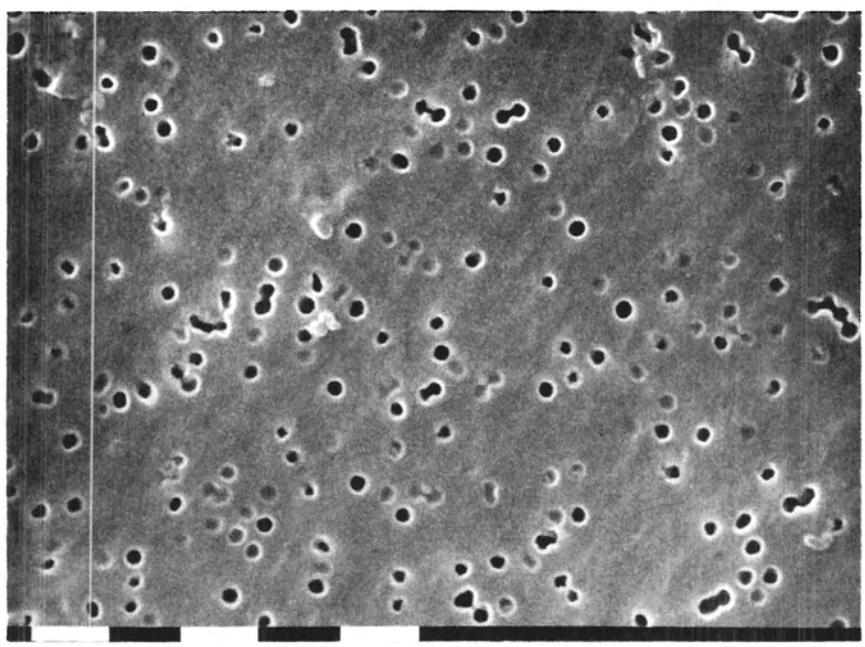


Fig. F.6d Same as c, but 10,000X magnification, bar scale is 1 μm.

these photographs only yielded a sulphur peak. This indicated that these particles, which represent the majority of the mass in these photographs, are composed primarily of light elements (at wt. <11). This suggests carbonaceous, organic, or secondary (such as $(NH_4)_2\ SO_4$) aerosol particles. The bright particles in these photographs are siliceous. This is in accordance with the observations of others who have shown that sulphate, organic and carbonaceous aerosol particles represent the majority of the mass of the total arctic aerosol at Barrow (Rahn and Heidam, 1981).

F.2a Cloud condensation nuclei

As a result of the higher aerosol loading during the winter at Barrow, the CCN concentrations are also higher than those of the summer. The CCN are in general 4-5 times higher in concentration at 0.2% SS during the winter than during the summer. Concentrations at this supersaturation were stable, having an average value of 150 cm^{-3} with a range of 125 cm^{-3} to 175 cm^{-3}. The data in Table 5.2 show the CCN to be significantly correlated with the pollutant tracers V and $SO_4^=$ as well as the CN concentrations. There is no relationship with the other tracers. Analysis for homogeneity of variance between CCN and V and $SO_4^=$ does not prove to be significant. Close scrutiny of the curves for these aerosol components in Fig. F.4 shos that the CCN had a relatively small variance in comparision to V and $SO_4^=$, but that their variations were coherent. This is not true of the other aerosol tracers. These pollutant tracers are representative of the long range transport of aerosol particles, both secondary and primary, from mid-latitude regions. The CCN are most closely related to the $SO_4^=$, most likely in the form of $(NH_4)_2SO_4$ or H_2SO_4. The 55% variance of the CCN explained by CN concentrations is

not indicative of local fresh emissions as CCN sources. Rather, considering the consistent NE winds, there was no possibility of local human activity having an influence on the aerosol. The reason for the close covariance is that the total aerosol is primarily composed of hygroscopic material, as suggested above, and that these aerosol particles are activated as CCN. Comparing the summer and winter study results, the CCN at 0.2% SS comprise 70% of the total CN concentration during the winter, and only 15% during the summer, on the average. Note that during the latter half of the winter study, when the arctic airmass was present along with the anthropogenic aerosol, the CN and CCN concentrations are the same. This is strong evidence of the secondary hygroscopic nature of the total aerosol present during the winter at Barrow. It is evident the majority of the CCN at Barrow during the winter are of anthropogenic origin.

F.2b Ice nuclei

The IN concentrations at $-20^{o}C$ also had a relatively small degree of variability. Average concentrations were 47 m^{-3} with a variation within a factor of two of the average. However, Table 5.2 shows the IN to be best correlated with the pollutant aerosol particle tracer V. This is indicative of the dependence of the IN on the primary pollutant aerosol. The dependence of the IN on marine Na has disappeared. This corresponds to the lack of open ocean water in the vicinity at this time of year. The marine aerosol component is still present, and in fact at a higher concentration than during the summer. As discussed previously, this has been attributed to the ablation of sea ice. This suggests the possible marine component of IN during the summer may be related to

biological activity, which is essentially absent during the winter due to the short solar day.

In spite of the presence of greater concentrations of all the aerosol tracers and the overall mass loading of the aerosol in general, as evidenced by comparison of the SEM photographs in Figures F.6 and F.3, the average IN concentrations are lower during the winter than during the summer. This shows that although the pollutant aerosol particles are primarily responsible for the IN concentrations during the winter, they are not as efficient as IN as the natural aerosol particles present during the summer. Additionally, the lower IN conentrations of winter may be a result of 'snowout' along the transport path in the cold Arctic. Temperatures were observed to be between -24°C and -32°C, below the activation temperatures used for the determination of IN concentrations in the laboratory. The temperatures aloft were 10°C to 20°C warmer. What significance the preferential removal of IN by activation might have relative to a possible 'poisoning' of IN by the higher concentrations of pollutants is not known. Certainly both effects are playing a role as discussed in Chapter 6.

F.3 Seasonal Differences in the Cloud-Active Aerosol

Figure F.7 presents the mean CCN spectra for both winter and summer at Barrow. Figures F.8 and F.9 depict the individual IN concentration for summer and winter with the mean spectra also plotted. High blanks during the processing of the IN filters at -28°C, caused the low IN concentratons. This was, in general, true for all the arctic sites. The data at -28°C are therefore questionable. A best estimate of the IN concentration at -28°C would be an extrapolation of the spectra from the other three temperatures.

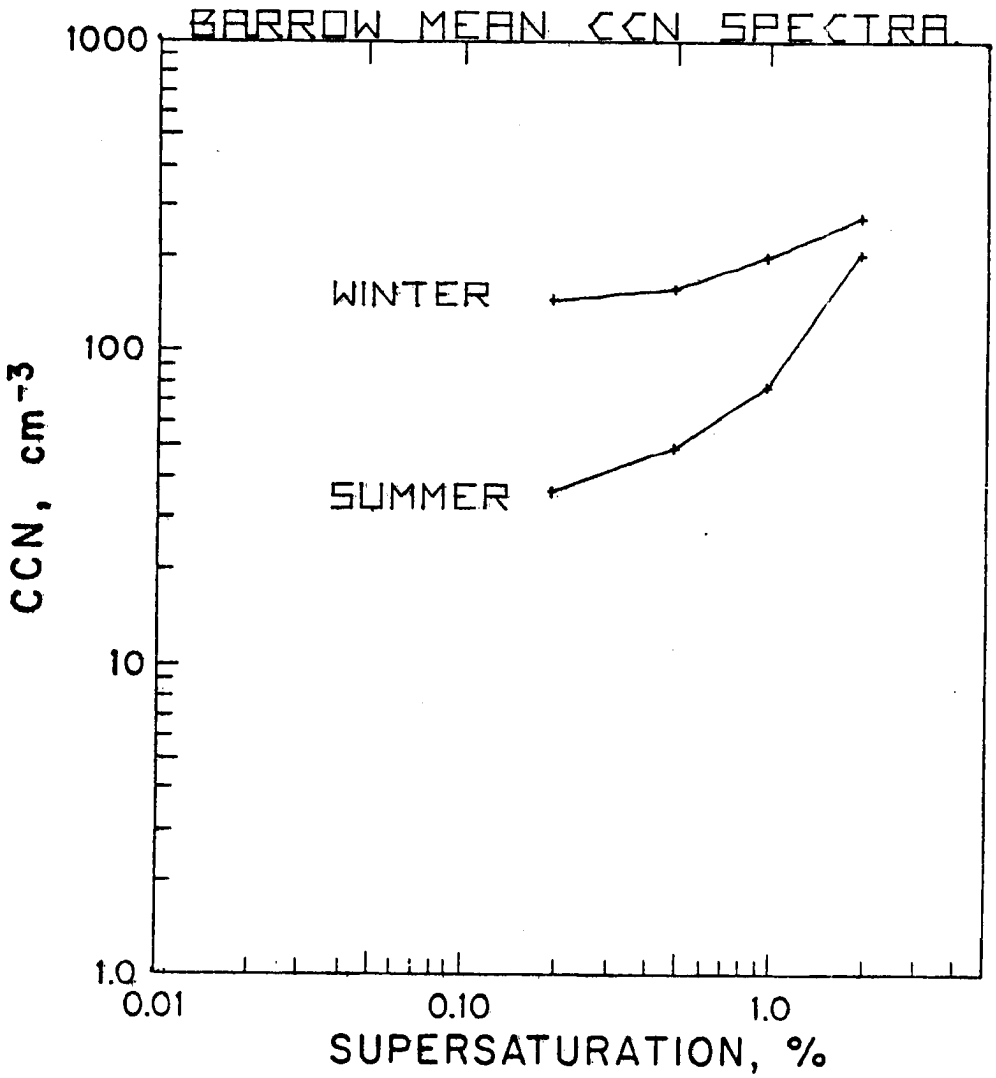


Fig. F.7 Mean CCN spectra for Barrow, Alaska, USA, derived from eighteen spectra measured during summer, 27 July - 7 August, 1979 and ten spectra measured during winter, 23 March - 3 April, 1980.

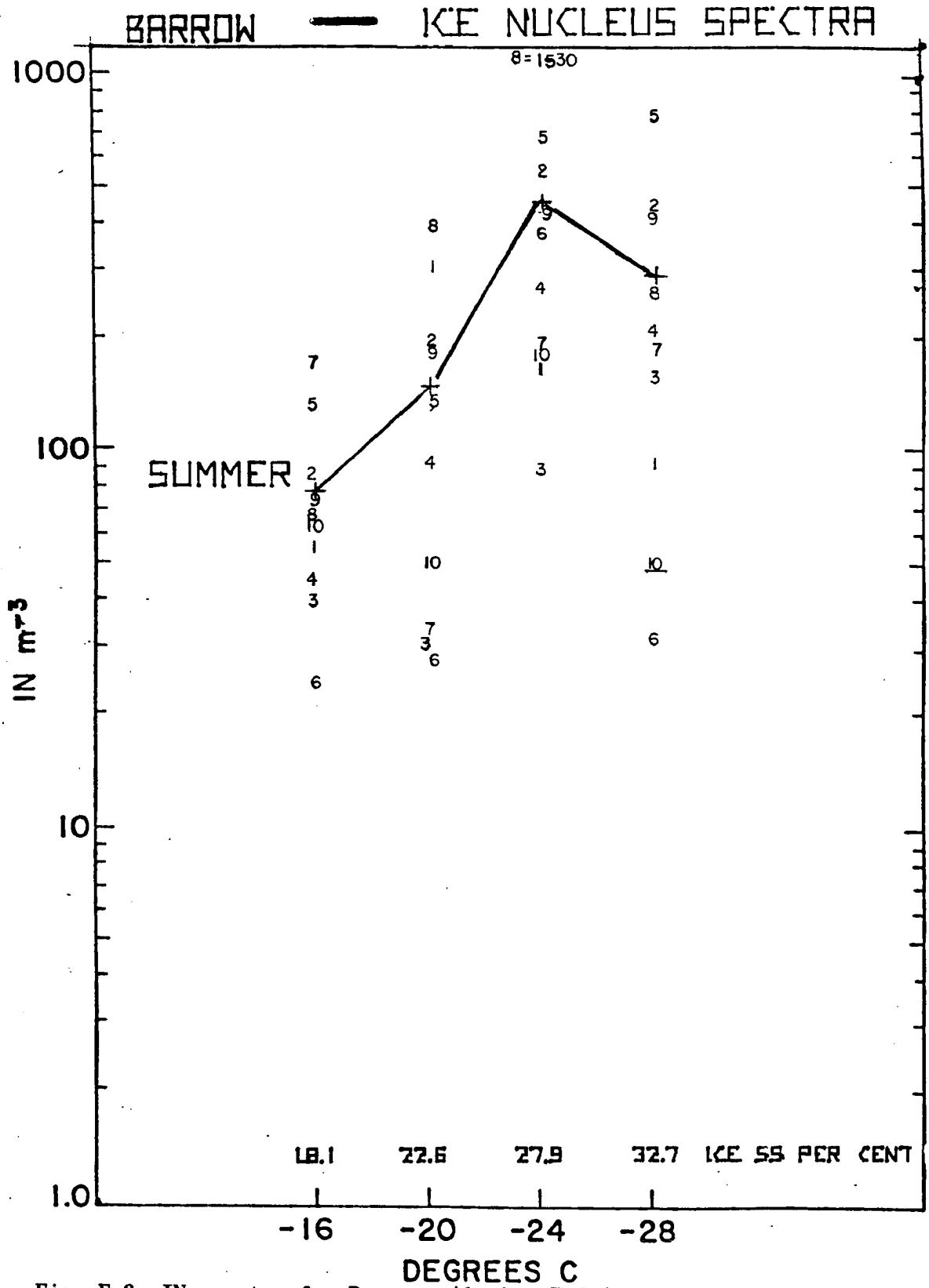


Fig. F.8 IN spectra for Barrow, Alaska, U.S.A. for the summer experiment, 24 July - 7 August 1979. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

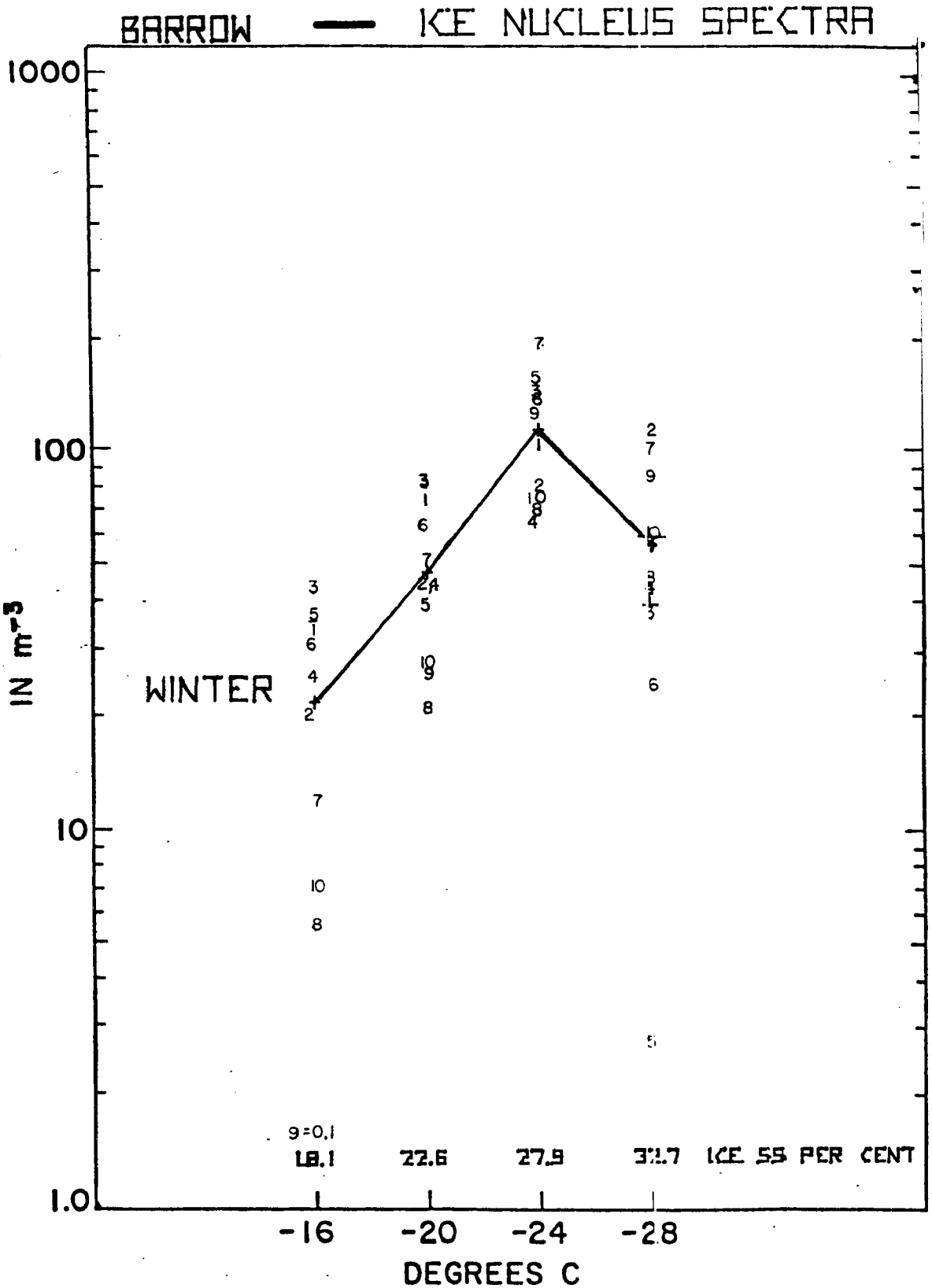


Fig. F.9 IN spectra for Barrow, Alaska, U.S.A. for the winter experiment, 24 March - 3 April 1980. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

There is clearly a difference between the concentrations of the cloud active aerosol of winter and summer. The winter CCN spectra are flatter and at higher concentrations. The greatest difference in the CCN concentration occurs at the lower supersaturations. This is in accordance with the observation of Arctic Haze during the winter-spring and not during the summer. Extrapolation of the CCN spectra to lower humidities approaching the region of activity of haze forming nuclei, provides an estimate of the difference in the magnitude of the concentrations of the nuclei responsible for Arctic Haze. The IN spectra show the opposite change in IN concentrations from summer to winter. Tables 5.5 and 5.4 show how these seasonal CCN and IN differences compare with the seasonal differences in the pollutant aerosol particle tracers. The factor of 4 increase in CCN and the factor of 3 decrease in IN occur at the same time there is a factor of about 40 increase in the pollutant aerosol. Combining the meteorology with these observations, the pollutant aerosol seasonal differences are shown to be a major factor in determining the seasonal differences in the CCN and possibly in the IN.

F.4 Sources of Cloud-Active Aerosol

From the discussions of the winter and summer aerosol experiments at Barrow, Alaska, it is apparent that the long-range transport of pollutant aerosol plays a significant role in determining the cloud-active aerosol concentrations. When this pollutant aerosol is present during the winter, both the CCN and IN correlate with its variation from day to day. When the pollutant aerosol is not present during the summer, natural sources of CCN and IN, as a result, are more important. Concentrations of CCN respond positively both on a seasonal and daily

basis to the pollutant aerosol. Thus the pollutant aerosol, which is present as a result of long range transport from mid-latitude sources, is a strong source of CCN at this site. Concentrations of IN respond negatively to the seasonal dependence of the pollutant aerosol. Concentrations of IN of natural origin during the summer are higher than thos of anthropogenic origin during the winter. Summer IN appear to have a marine, possibly biogenic, origin. Winter IN, although from anthropogenic sources, are fewer in number as a result of a combination of pollutant interaction with the IN resulting in a deactivation of 'poisoning' of the IN and a selective removal of IN during the winter. Anthropogenic sources of IN and CCN are important at this site during the winter. Natural sources of IN and CCN dominate during the summer.

It has been shown that the important sources of IN and CCN are advected to Barrow from the north. These include both the natural and anthropogenic sources. This eliminates the transport over the North Pacific of natural and anthropogenic aerosol from oceanic and continental regions as a major source of cloud active aersol at Barrow. Transport along the North Pacific pathway certainly occurs as shown by, Rahn et al. (1977b). However, this is one of the most stormy regions in the northern hemisphere. Removal processes along this pathway are efficient in cleansing the atmosphere of any aerosol from sources far upwind. These removal processes do not permit the survival of the aerosol particles along this pathyway.

APPENDIX G

DETAILED FIELD EXPERIMENT DISCUSSION
Poker Flat, Alaska, USA

This site was chosen to determine the importance of the North Pacific transport pathway of aerosol particles to the Arctic. Situated in the interior of Alaska on a ridge top at an elevation of 490 m, the site is representative of the aerosol climatology in central Alaska. The site is located aproximately 350 km south of the Brooks Range. This places the site about 600 km south of the Barrow site. The nearest source of local contamination is the city of Fairbanks, Alaska, about 25 km to the south.

G.1 Summer Study

The summer study period extended from 17-25 July, 1979. Nine, 24-hour sample sets were collected and analyzed. The results of the bulk-aerosol trace-element composition are given in Table 5, Appendix A. Figure G.1 is a plot of the day-to-day variation of the aerosol tracer components of the total aerosol and the CCN and IN concentrations at 0.2% SS and $-20^{\circ}C$, 1% WSS respectively. Figures G.2a-d are the isohypse analyses for the 850 mb and 700 mb pressure surfaces for the study period. The same notation applies to these analyses as discussed previously.

Being an interior continental site, the only one of its kind in this report, the measurable impact of marine aerosol is minimal. Marine Na and I are both near their detection limit. The natural continental

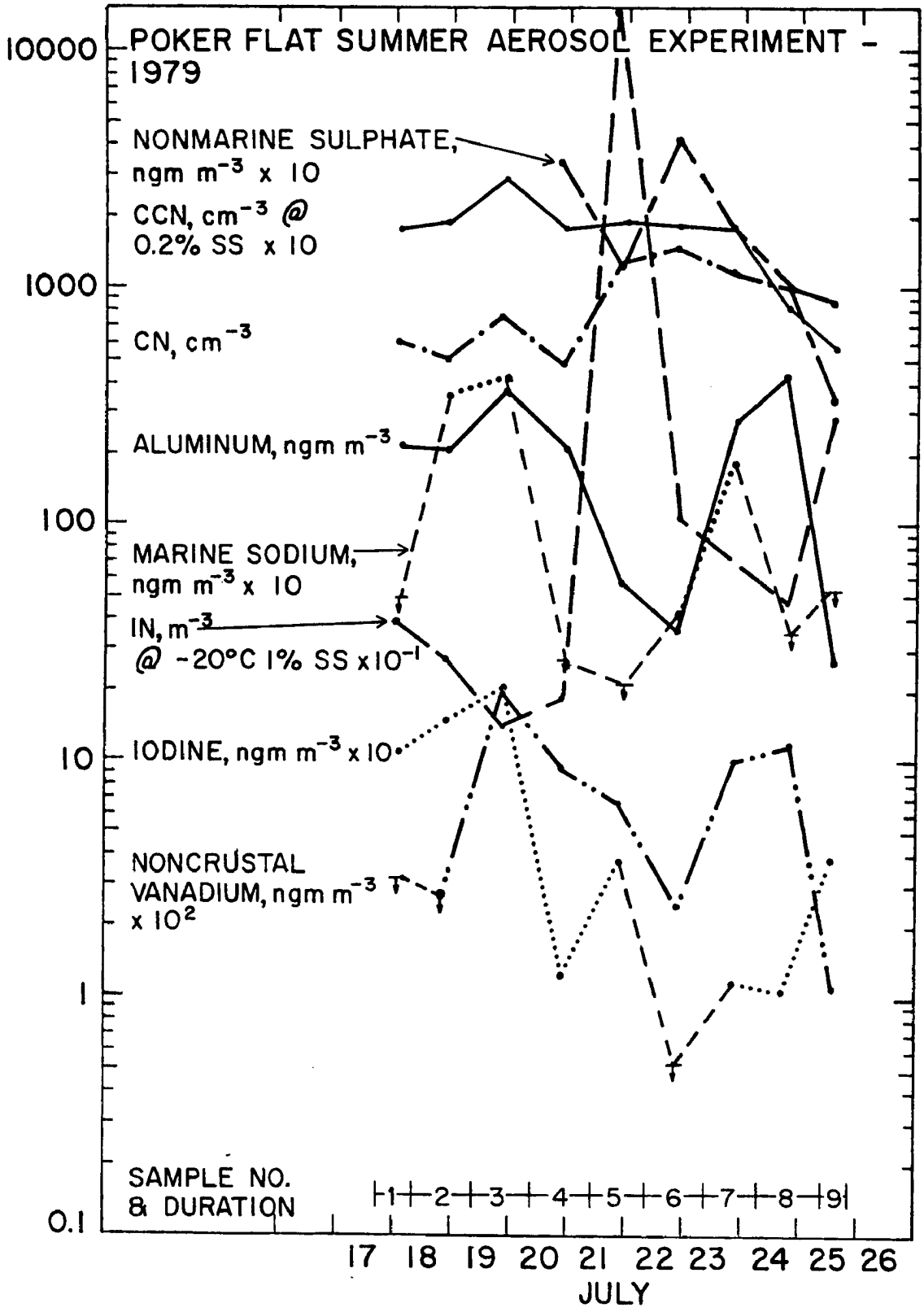


Fig. G.1 Poker Flat, Alaska, USA summer aerosol experiment, July 1979.

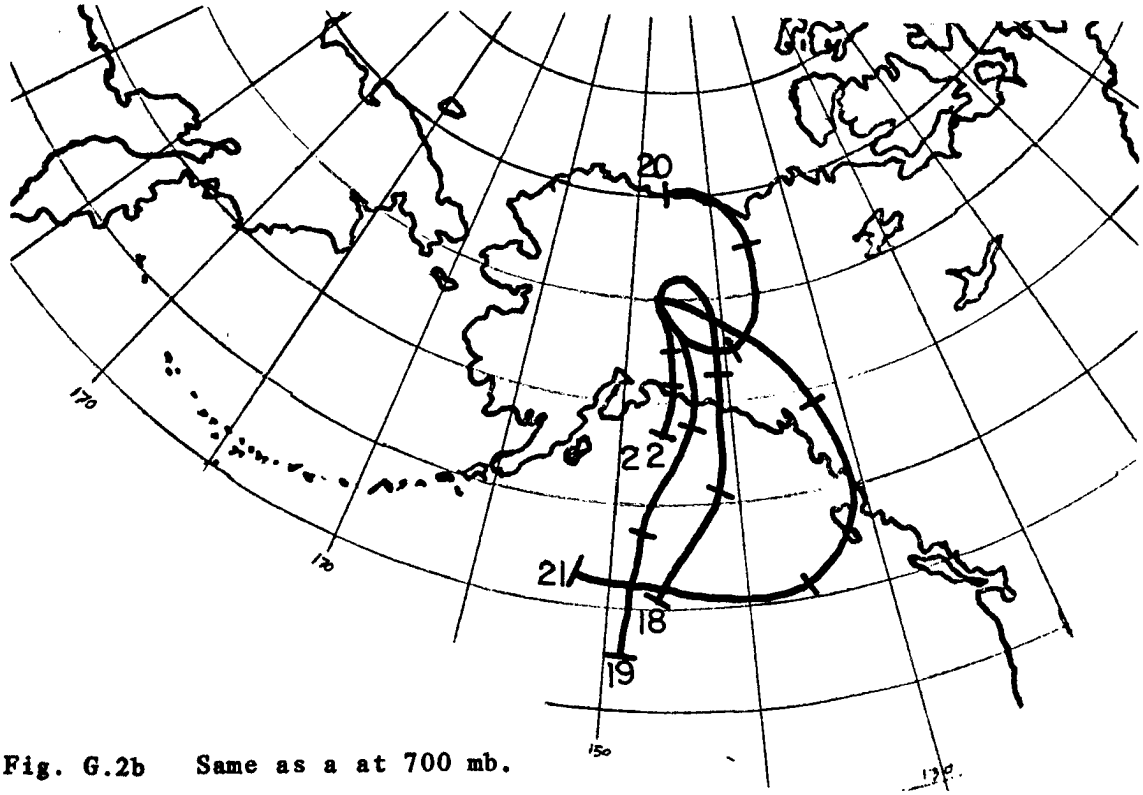


Fig. G.2b Same as a at 700 mb.

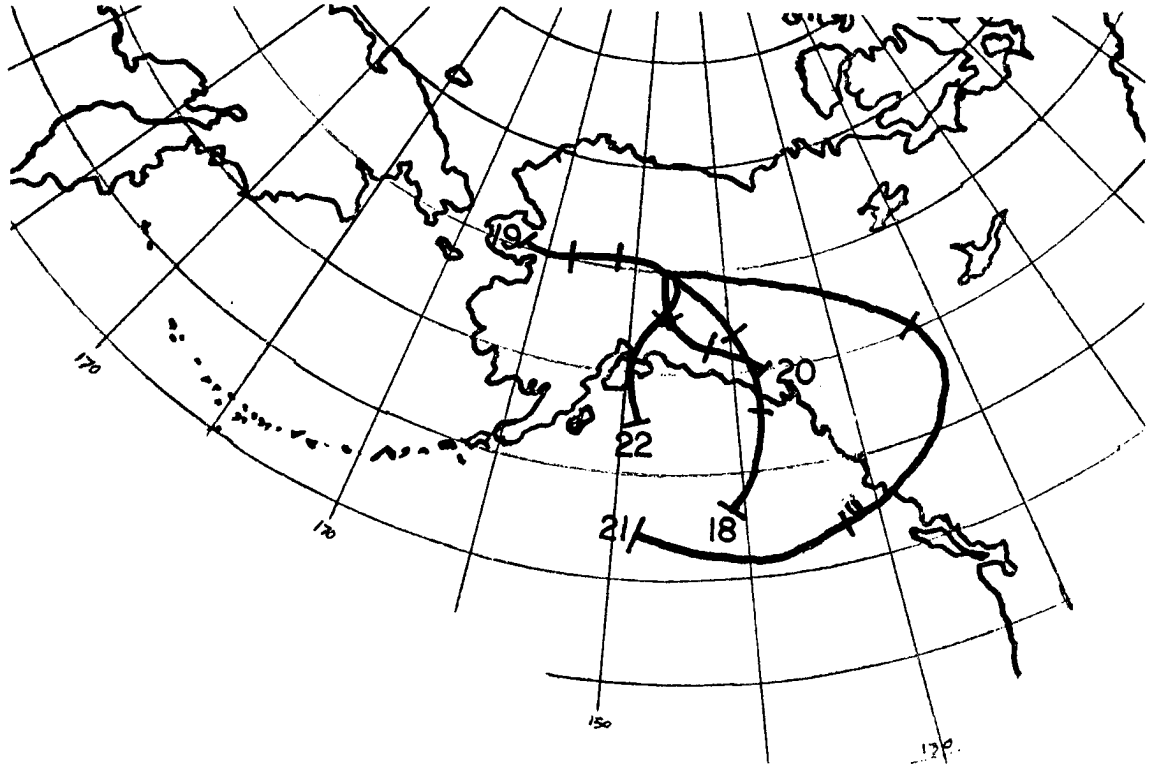


Fig. G.2a Poker Flat 850 mb isohypse analyses, 18 - 22 July, 1979, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

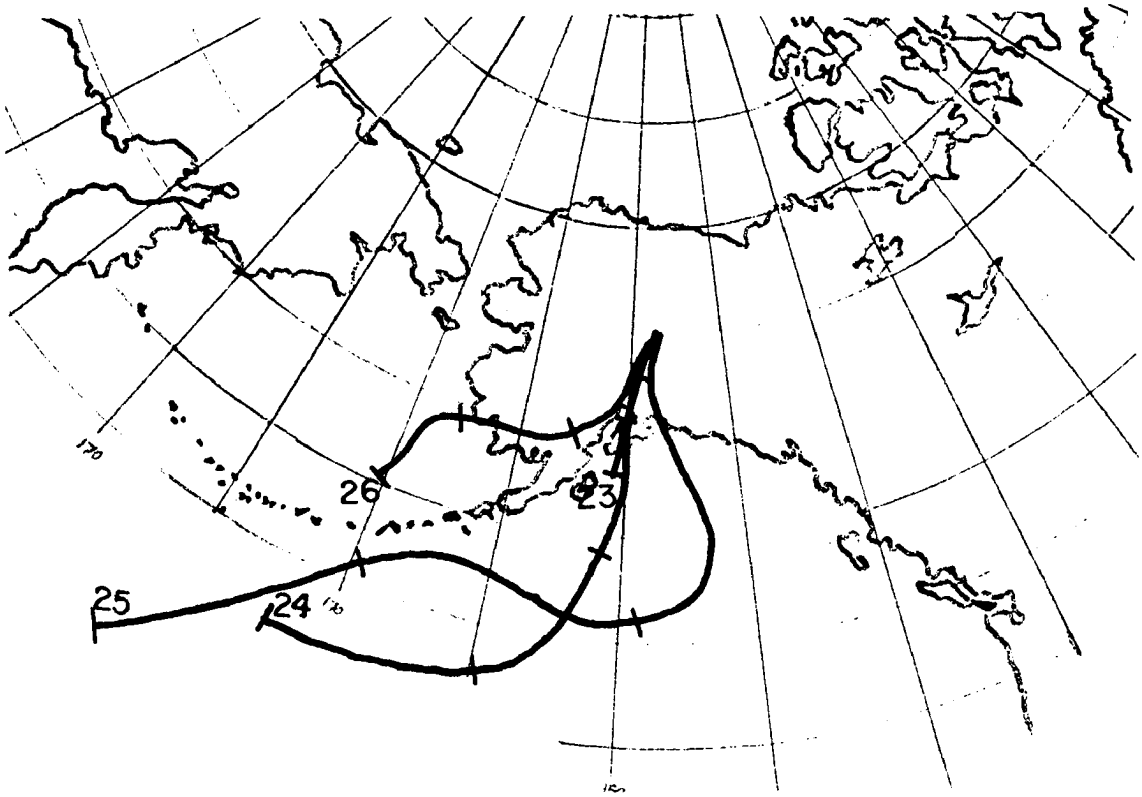


Fig. G.2d Same as a at 700 mb, 23 - 26 July, 1979.

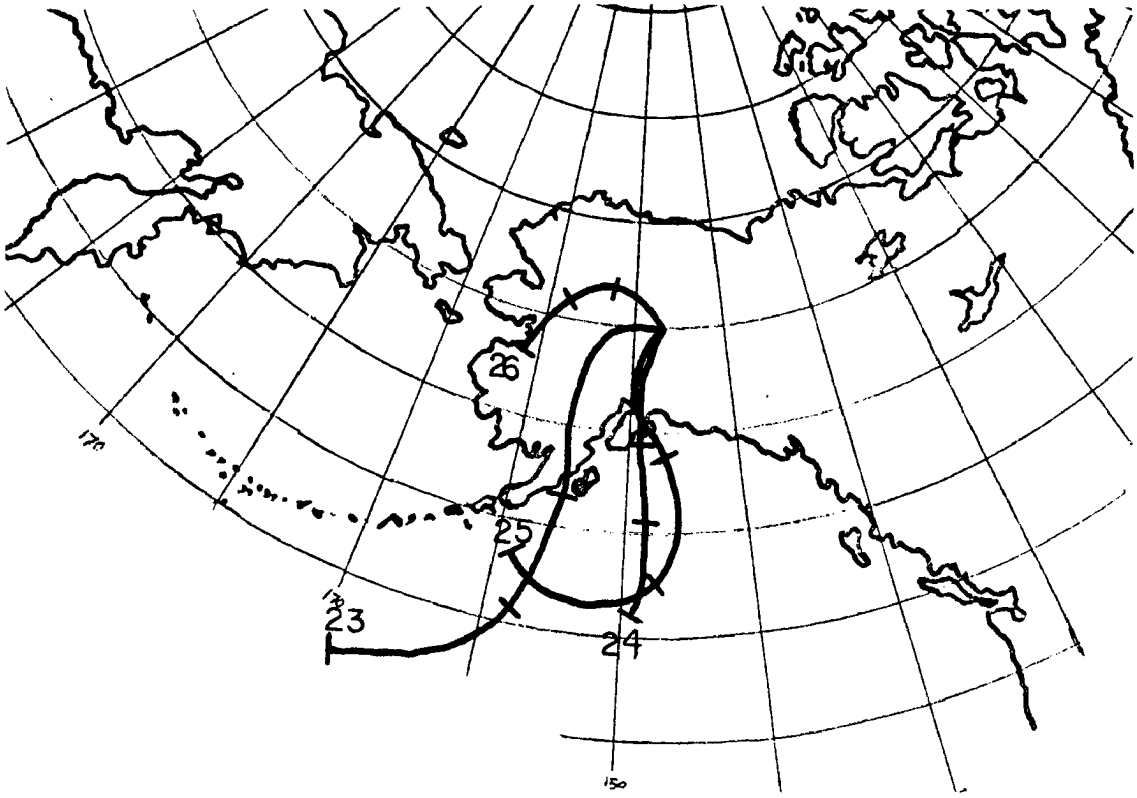


Fig. G.2c Same as a at 850 mb, 23 - 26 July, 1979.

tracer, Al is over an order of magnitude greater in concentration in Poker Flat than at Barrow during the summer season. Greater biogenic activity, associated with the forested region, is probably responsible for the higher concentrations of non-marine $SO_4^=$ at this location in comparison to Barrow. Noncrustal V concentrations are very low. Poker Flat, located at 65°N, is not considered to be a high-arctic site. Rather, it is a mediate site as a result of its location, south of the Arctic Circle.

During the entire summer study period, the North Pacific Ocean was the origin of airmasses which were advected to this site. The circulation was relatively weak. The airmasses that reached Poker Flat during this period were modified-maritime. That is, their source region was the Gulf of Alaska but they were advected over approximately 500 km of interior Alaska. The weather during the period was characterized by low wind speeds and showers associated with non-frontal convection. The aerosol which dominated the sampling period should be representative of an interior Alaskan airmass.

The low concentrations of the elements as depicted in Figure G.1 and Table 5 in Appendix A are reflected in the SEM photographs in Figure G.3a-f. These photographs are of the aerosol from samples 3, 5 and 9. Sample 5 will be discussed further under the section on IN. The volumes sampled that these photomicrographs represent are one fourth the volumes that were collected for the Barrow photographs. The areas are representative of approximately one and ten cm^3 for the 10,000X and 3100X photomicrographs respectively.

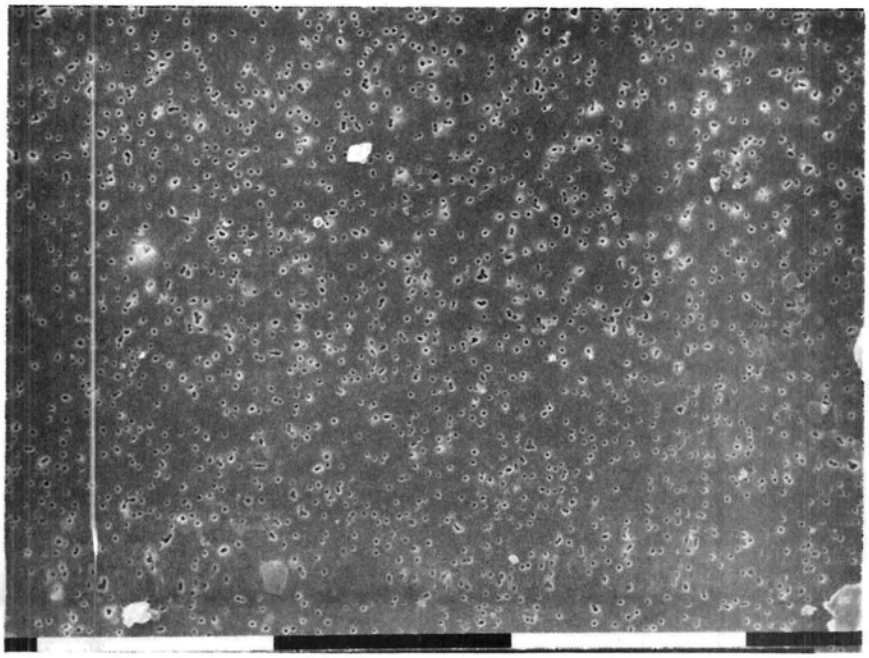


Fig. G.3a Scanning electron microscope photographs of aerosol particles collected at Poker Flat during summer on 0.2 μm diameter pore size Nuclepore filters from sample #3, 3100X magnification, bar scale is 10 μm.

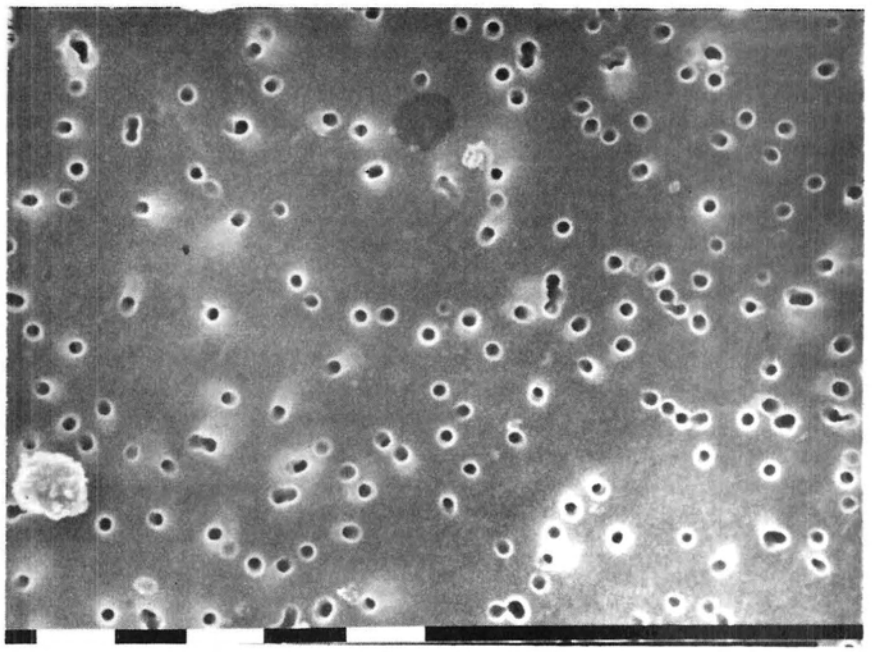


Fig. G.3b Same as a, but 10,000X magnification, bar scale is 1 μm.

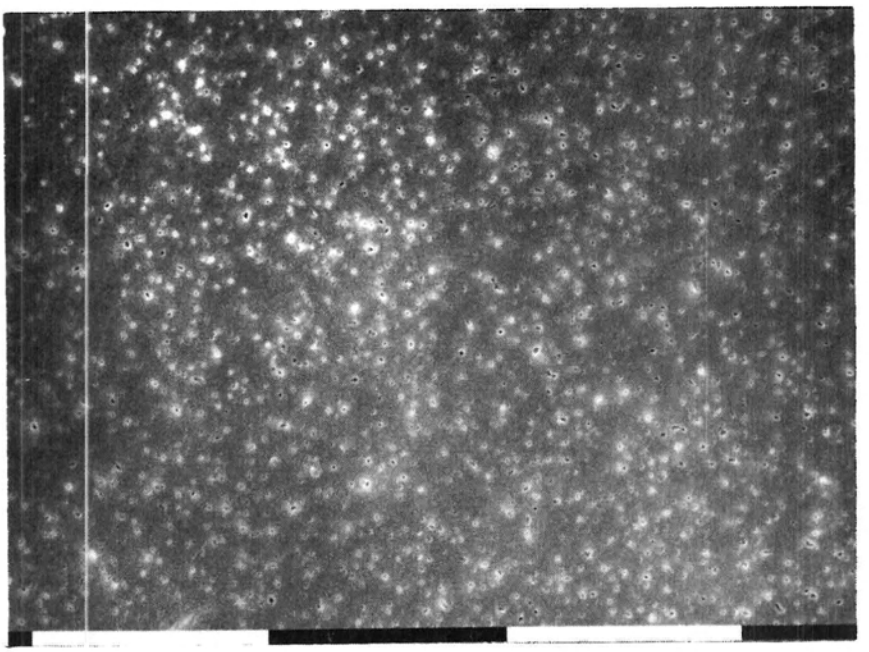


Fig. G.3c Same as a but for sample #5.

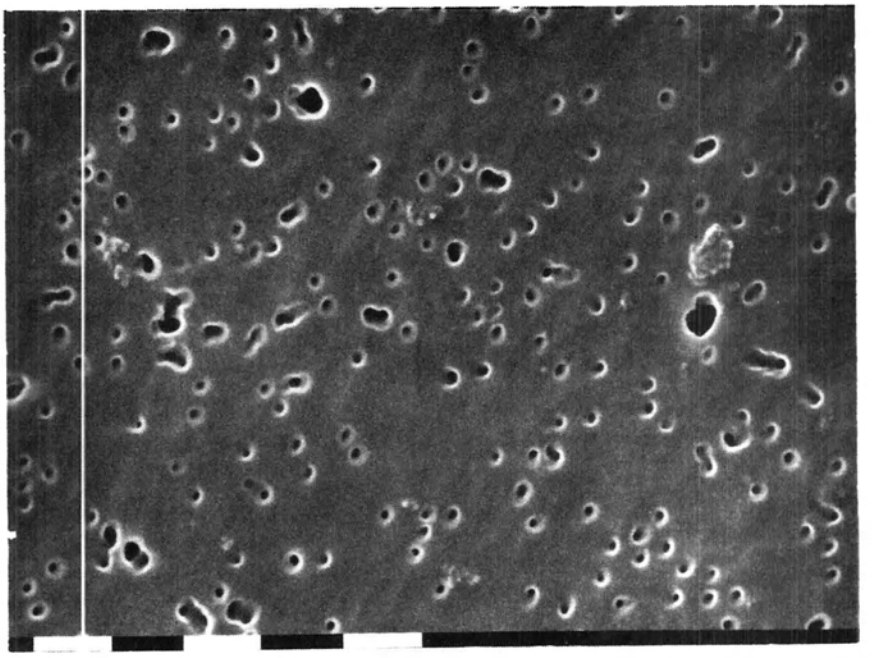


Fig. G.3d Same as c but 10,000X magnification, bar scale is 1 μm.

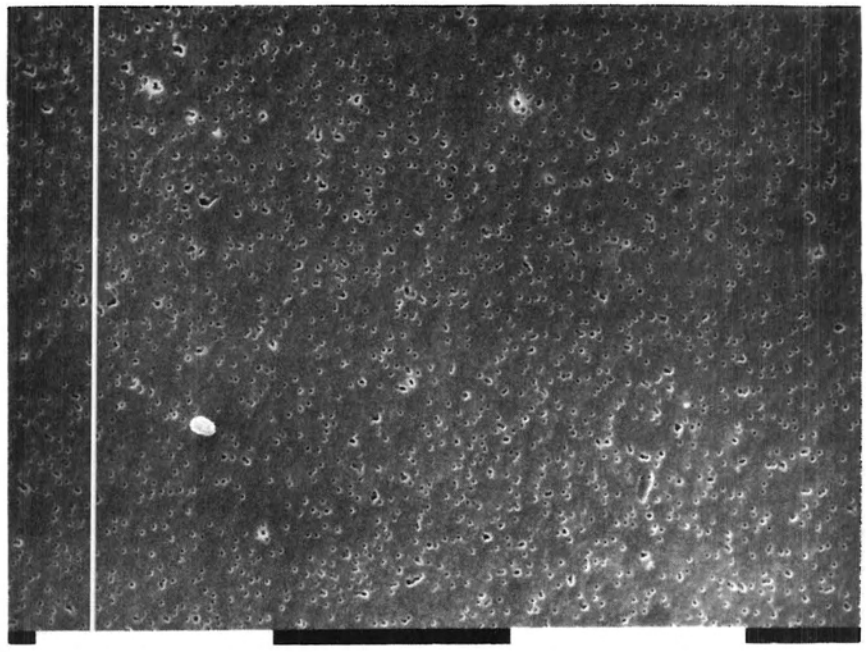


Fig. G.3e **Same as a but for sample #9.**

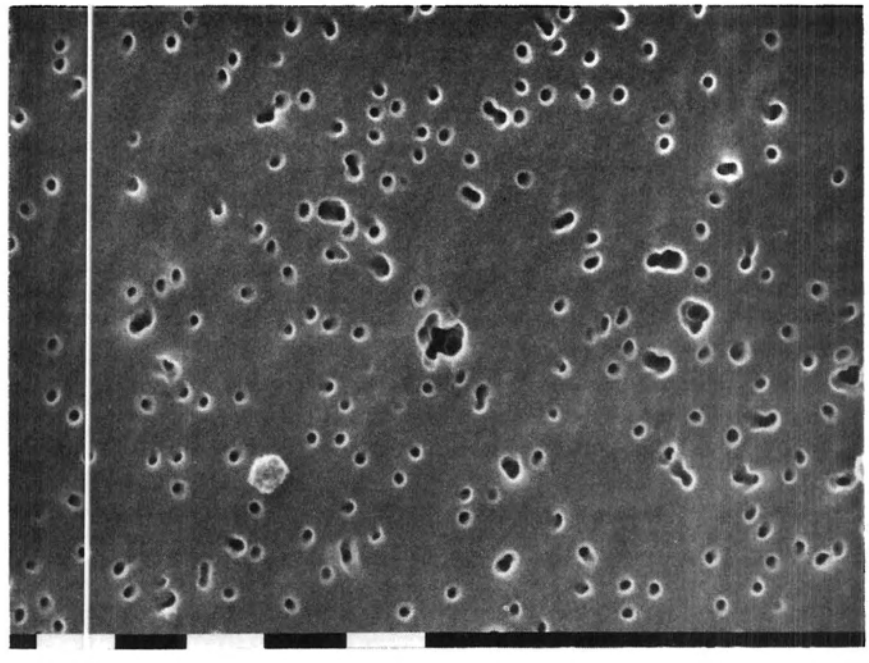


Fig. G.3f **Same as e but 10,000X magnification, bar scale is 1 μm.**

G.1a Cloud condensation nuclei

Although the concentrations of CCN are 4-5 times greater than those at Barrow during the summer, little can be deducted about their origin because of the low concentrations of the aerosol elemental tracers. None of the data in Table 5.2 explaining the per cent variance of the CCN at Poker Flat during summer are significant. Figure G.1 shows the CCN concentrations were higher during the earlier part of the study period than during the latter part. The variation of the CCN is smoother than that of any of the aerosol particle elemental components measured. The sluggish air motion which dominated the period is shown by the short length of the isohypses in Figure G.2. From this, and the low concentrations of maritime and pollutant indicators, the aerosol particles present, and therefore the CCN, are likely of local origin. Since no correlation occurred with any of the elemental tracers, the CCN may be related to some unmeasured component of the aerosol.

G.1b Ice nuclei

The IN concentrations undergo a large variation in concentration, yet do not correlate with any of the measured aerosol parameters. This can be seen in Figure G.1 and Table 5.2. Sample 5 stands out as an unusually high concentration of IN. The sample was characterized by low cloudiness and light rain. Inspection of the SEM photograph in Figure G.3 for this sample reveals many more small particles approximately 0.1 μm in radius than were present in samples 3 or 9. The origin and composition of these particles is unknown. The concentrations of IN other than sample 5 are approximately fives times greater than those measured at Barrow. This is about the same difference as that found for the CCN concentrations. Based upon the meteorology and aerosol

composition, the IN measured are most likely of local origin and possibly related to a component of the aerosol which was not measured. Marine and pollution sources can effectively be ruled out as potential sources because of the low to undetectable concentrations of their aerosol tracers.

G.2 Winter study

The winter study period extended from 4-13 April, 1980. Ten sample sets were collected and analyzed. The results of the bulk-aerosol trace-element composition is summarized in Table 6 of Appendix A. The day-to-day variation of the aerosol source tracer components are plotted in Figure G.4 along with the CCN at 0.2% SS and IN at -20^{o}C and 1% WSS. Figures G.5a-d are the isohypse analyses for this period and site for the 850 mb and 700 mb levels. The same notation is used as for previous isohypse analyses. SEM photomicrographs of aerosol from samples 7 and 9 are presented in Figures G.6a-d.

The period of study is early spring. A true mid-winter period was not chosen because of the intent to perform the field work at this interior Alaskan site to correspond as closely as possible to the time of the study at Barrow, Alaska where the peak in pollutant aerosol is observed at this time of the year. Comparing Figures G.1 and G.4, winter concentrations of the aerosol elementral tracers at Poker Flat during the winter are, in general, higher and more coherent than during the summer. The aerosol component concentrations lie between the winter and summer values of the high arctic site, Barrow. The aerosol components reflect a general mix of marine, continental and pollution aerosol sources. This is a result of the site lying in a continental location approximately midway between the pollution aerosol of the

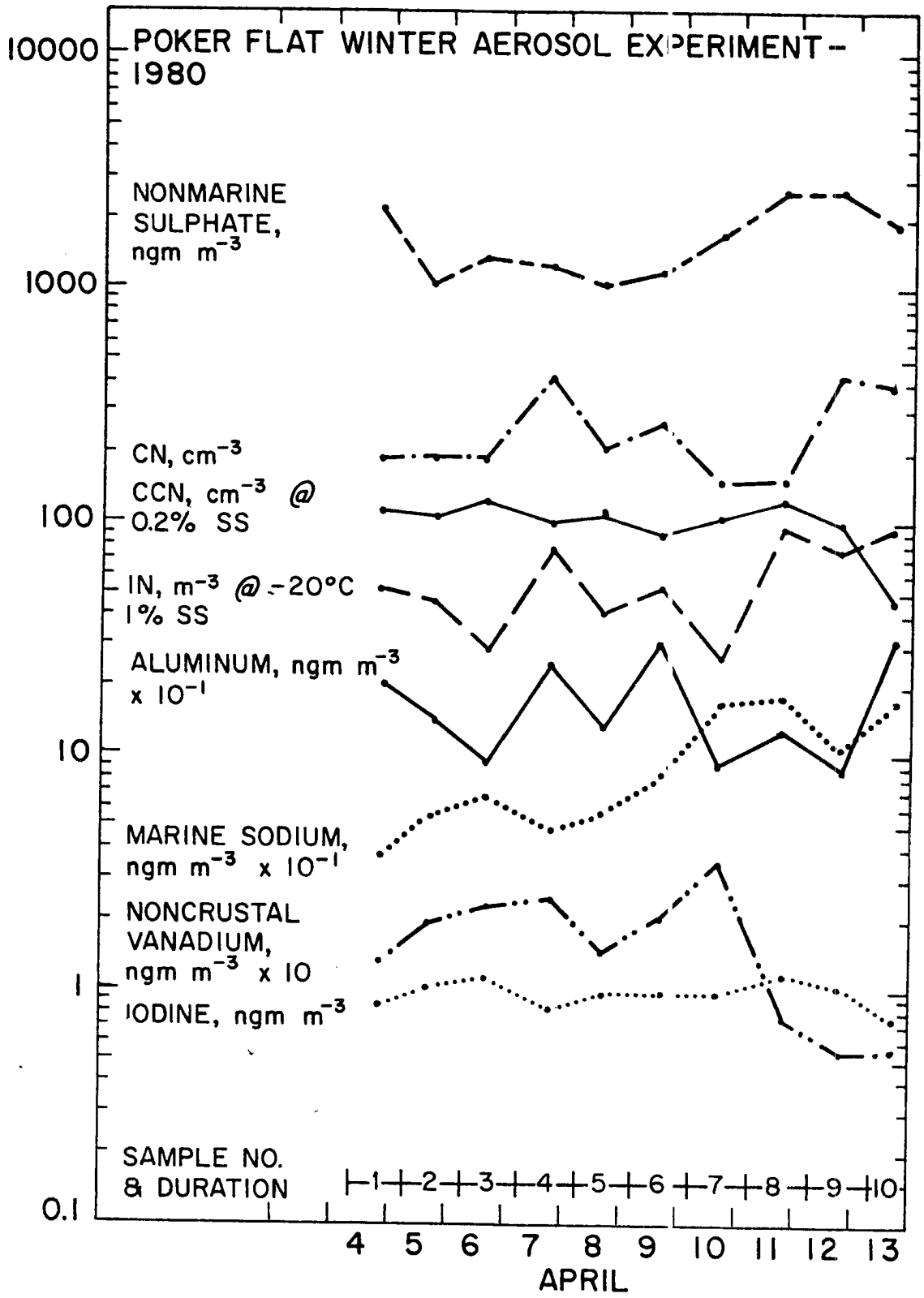


Fig. G.4 Poker Flat, Alaska, USA winter aerosol experiment, April 1980.

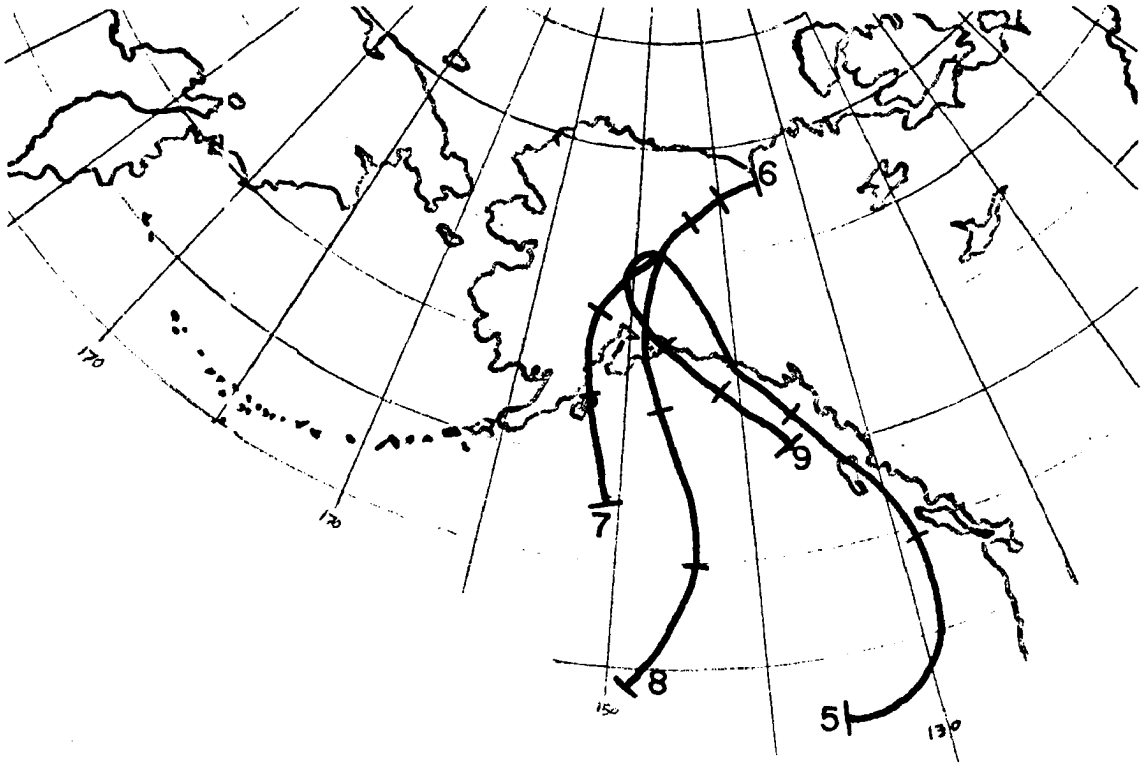


Fig. G.5b Same as a at 700 mb.

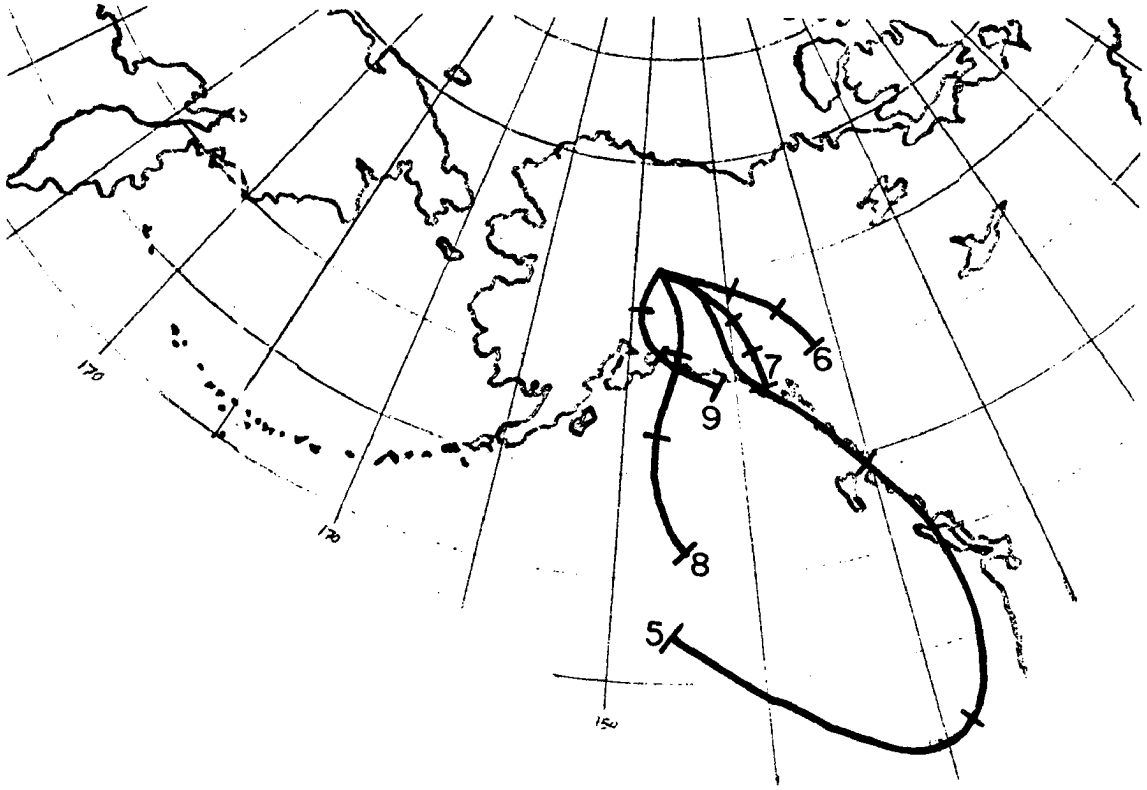


Fig. G.5a Poker Flat 850 mb isohypse analyses, 5 - 9 April, 1980, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

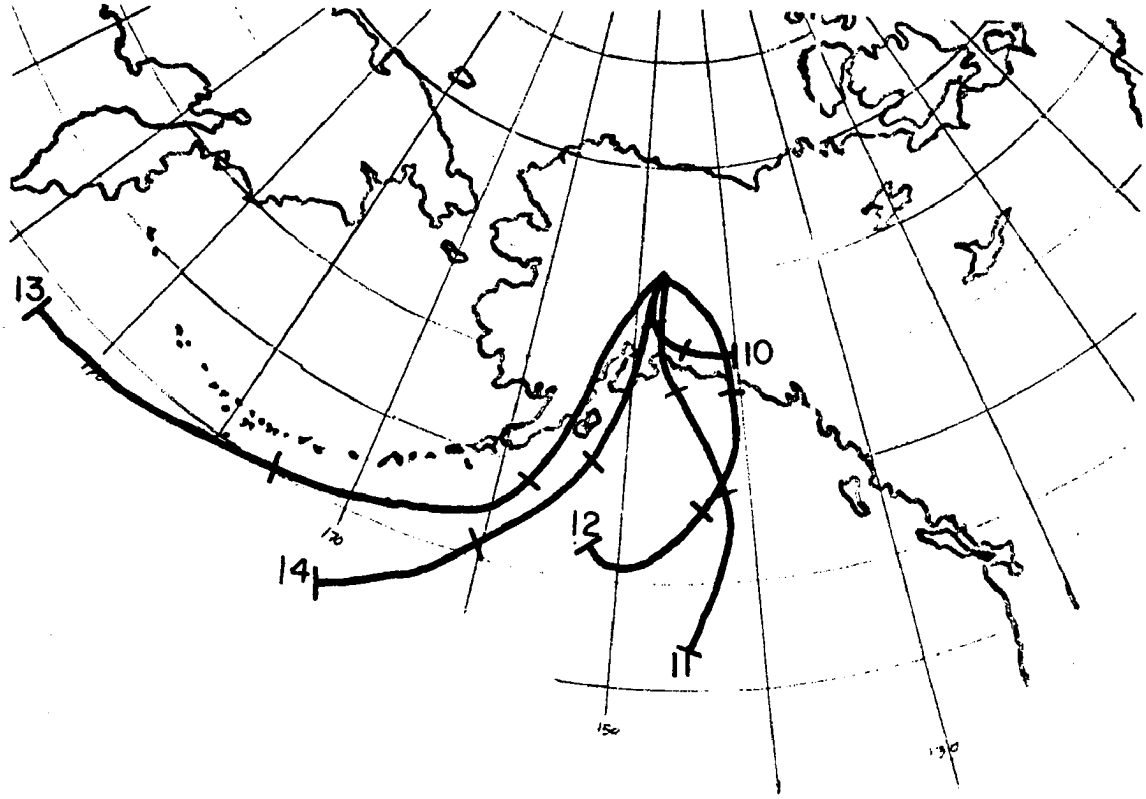


Fig. G.5d Same as a at 700 mb, 10 -14 April, 1980.

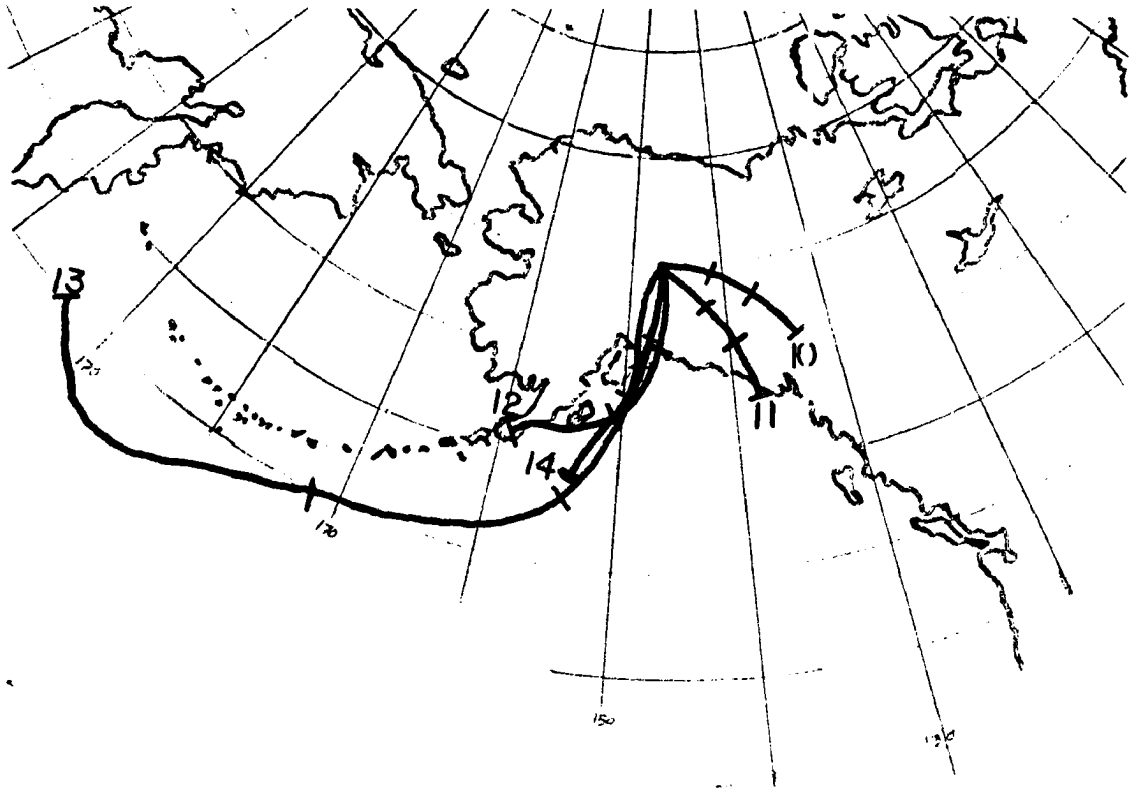


Fig. G.5c Same as a at 850 mb, 10 - 14 April, 1980.

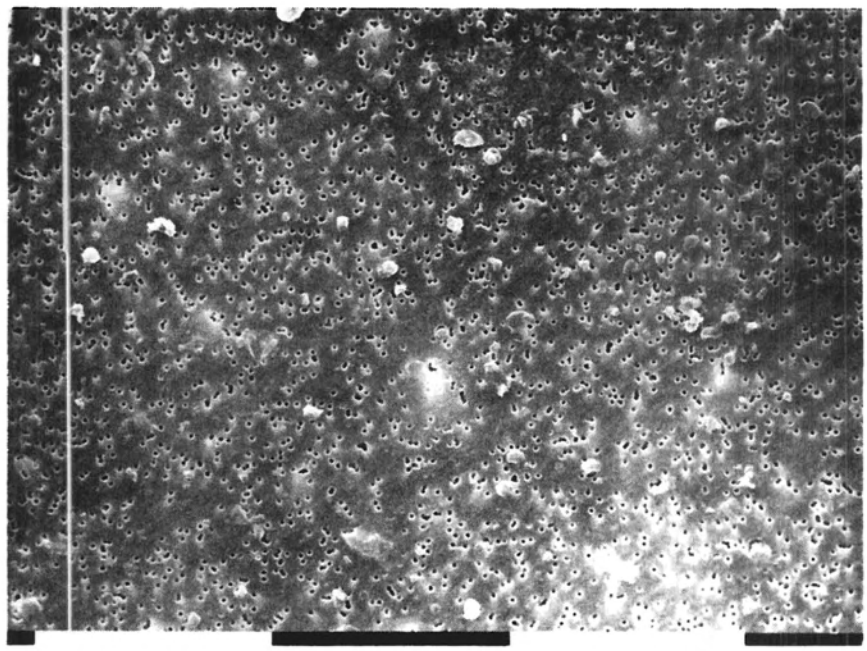


Fig. G.6a Scanning electron microscope photographs of aerosol particles collected at Poker Flat during winter on 0.2 μm diameter pore size Nuclepore filters from sample #9, 3100X magnification, bar scale is 10 μm.

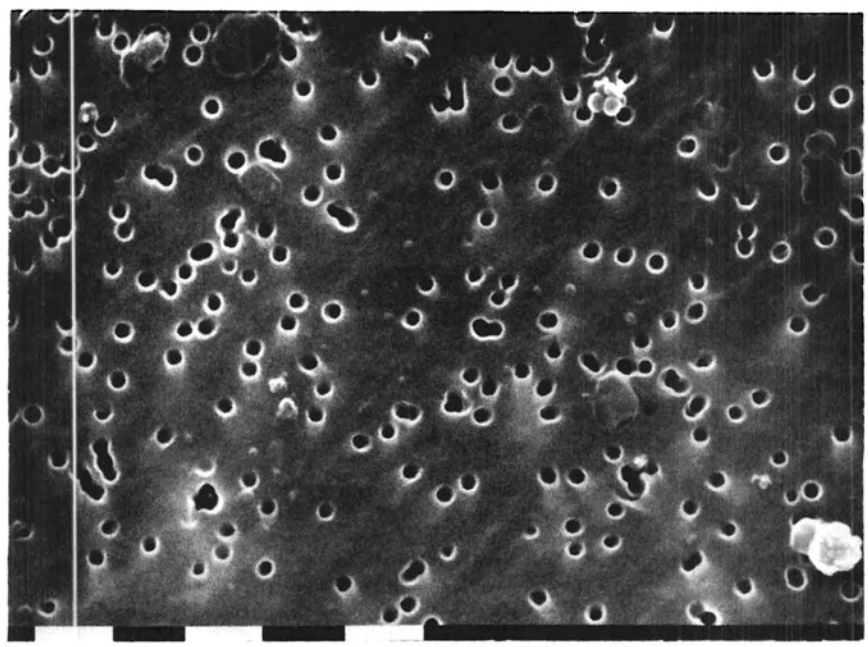


Fig. G.6b Same as a but 10,000X magnification, bar scale is 1 μm.

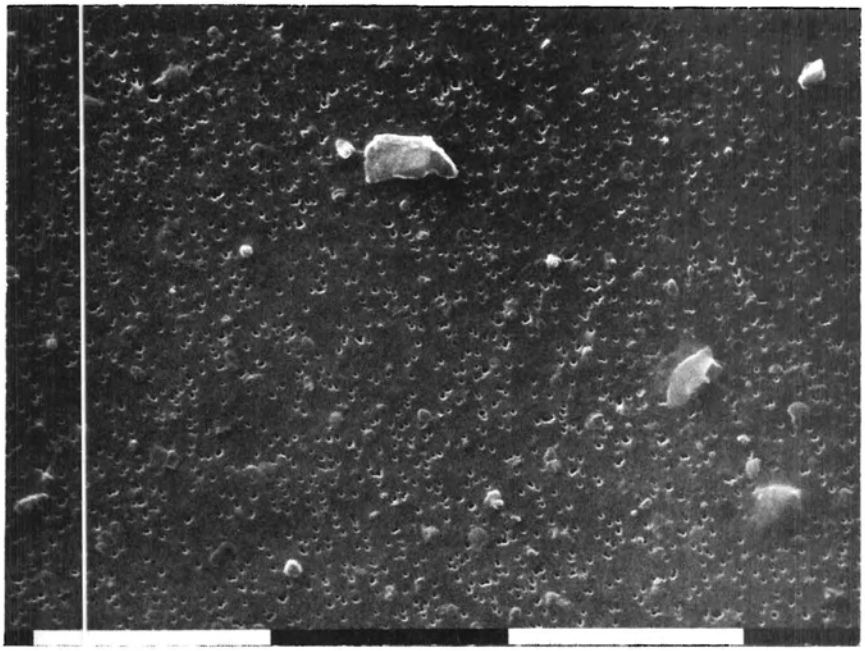


Fig. G.6c Same as a but for sample #9.

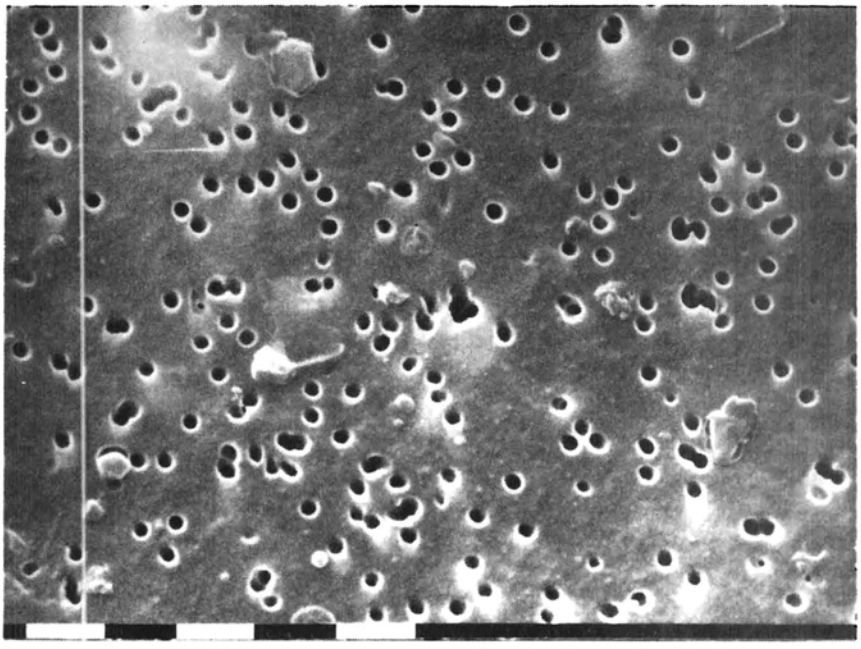


Fig. G.6d Same as c, but 10,000X magnification, bar scale is 1 μm.

arctic to the north and the marine aerosol of the Pacific Ocean to the south.

The meteorology of transport of this location is more complex than suggested by the isohypse analyses of Figure G.5. The isohypses show persistent flow from the southeast to southwest at the 850 mb and 700 mb levels. However, southerly flow only occurred at the surface during samples 4 and 9, with more vigorous southerly flow during sample 9. For all other samples, the winds were persistently out of the northeast. The coldest temperatures occurred during sample 7. The warmest conditions occurred during the periods of southerly winds. During samples 9 and 10 temperatures exceeded $0^{o}C$. A review of surface weather maps showed the weather at this time to be controlled by the surface Arctic high and Aleutian low pressure systems. Surface stations in northeast Alaska and northwest Canada were reporting northeasterly winds during much of the period. It is, therefore, likely that the dominant transport in the boundary layer was from the northeast, with perturbations on this flow as a result of northward movement of the Aleutian low, and subsequent flow from the south, reaching the surface.

The aerosol tracer composition reflects the meteorology. Considering that pulses of southerly flow occurred around samples 4 and 9, peaks in the CN concentration at these times represent the influence of the city of Fairbanks about 25 km to the south. The marine indicators Na and I suggest the incoming marine airmasses around the times of southerly flow at the surface. I shows little variation. However, marine Na generally increases during the period. The pollutant tracers are apparently responding to two sources since non-marine $SO_4^{=}$ obtains its highest concentrations during the strongest southerly flow

event and non-crustal V its lowest. The explanation is that there are coal-fired power plants in Fairbanks, sources of SO_2, a precursor to $SO_4^=$. However, there are no sources of V, which result from heavy crude oil combustion. The V concentrations peak during the cold, northeast flow situation. It is apparent the aerosol particles present during sample 7 are those which have been advected from the high arctic. Figure G.6 shows that, in general, the aerosol particles are of smaller size during the presence of aerosol from the north versus those from the south. Note the apparent fly ash spheres in Figure G.6b, collected during flow from the south. The concentrations of CN, V and $SO_4^=$ in sample 7 are all approximately the same as those measured at Barrow a week earlier (see Figure 5.12). Marine Na is also high at this time since the average concentrations of this component at Barrow were 6-7 times higher than those at Poker Flat. Thus, flow from both the northeast and the south can advect marine aerosol components to this site. The Al concentrations have a great deal of scatter, and average about seven times higher than those at Barrow, reflecting the continental nature of this site and land mass in all directions.

G.2a Cloud condensation nuclei

The concentrations of CCN at 0.2% SS varied little until the end of the period. Average values were 100 ± 20 cm^{-3}. Review of the percent variance of the CCN explained at Poker Flat during winter from the data in Table 5.2 suggests the CCN are not explained by local continental aerosol sources, using Al as an indicator. Secondary marine aerosol, as indicated by I, may play a role as a source for the CCN. The correlation with I may be a result of the fact that both components of the aerosol had the lowest variance of the sample set. The negative

correlation with Al is probably a good indication that sources of dust in the region are not important as CCN. The CCN undergo the greatest change in concentration during samples 8, 9 and 10. During this time, the surface wind direction was changing to south and increasing in speed and gustiness. The other component which responds to this change in weather conditions is the pollutant tracer V. This sensitive tracer of pollutant aerosol from the north suggests that these aerosols may play a role in determining a part of the CCN concentration of interior Alaska. This is a tentative conclusion based upon little data. Further studies over longer periods would be needed to determine if indeed this is the case.

G.2b Ice nuclei

The IN concentratons are more constant and much lower than those of the summer. The aerosol is, in general, higher in concentration for the components measured than during the summer. The data in Table 5.2 indicate no significant positive correlation with any of the aerosol components. There is a significant negative correlation with noncrustal V. As discussed previously, samples 8, 9 and 10 represent the flushing of the northern aerosol and airmass with winds from the south, which resulted in an increase in IN concentration. This is consistent with the observations of IN at Barrow. Concentrations of IN at Barrow during winter (the week prior to the measurements made at Poker Flat) averaged 20 percent less than at Poker Flat whereas concentrations of V were 4.5 times higher. The advection of this Arctic airmass from the north to Poker Flat would cause a decrease in IN and an increase in V, producing the inverse correlation between IN and V as shown in Table 5.2. The reverse of this is not true at Barrow because of the overwhelming

dominance of the polluted arctic airmass at all times. The IN at Barrow respond to the relative intensity of the polluted airmass, producing a positive correlation between IN and the pollutant tracer V. This is the result of only three measurements and may not, in general, be representative of the true relationship of IN and aerosol advected from the north. There is an indication of a slight positive relationship with the CN concentrations which have been shown to be associated with aerosol advected from the Fairbanks area. The other tracer data does not give any further evidence as to the possible significance of this relationship.

G.3 Seasonal Differences in Cloud Active Aerosol

Figure G.7 presents the seasonal mean CCN spectra for winter and summer at Poker Flat. Figure G.8 and G.9 depict the individual IN concentrations for summer and winter with the means spectra also plotted.o Data for $-28^{o}C$ are questionable due to large blank values during analysis at this temperature. The basic difference between the two seasons are the higher concentrations of summer versus those of winter. Tables 5.3 and 5.4 show that although the winter/summer ratio of the pollutant tracers are greater than one, the IN and CCN ratios are less than one. The cloud-active aerosol concentrations are a result of a source of aerosol which is present during the summer and not in the winter. The winter CCN spectra has the more shallow slope expected from a location affected by pollutants. The slope of the Barrow winter CCN spectra are shallower than the less polluted summer case. However, the absolute concentrations of CCN do not follow the change in the concentration of the pollutant tracers with season. The most important result is the obvious difference of the change in the cloud-active

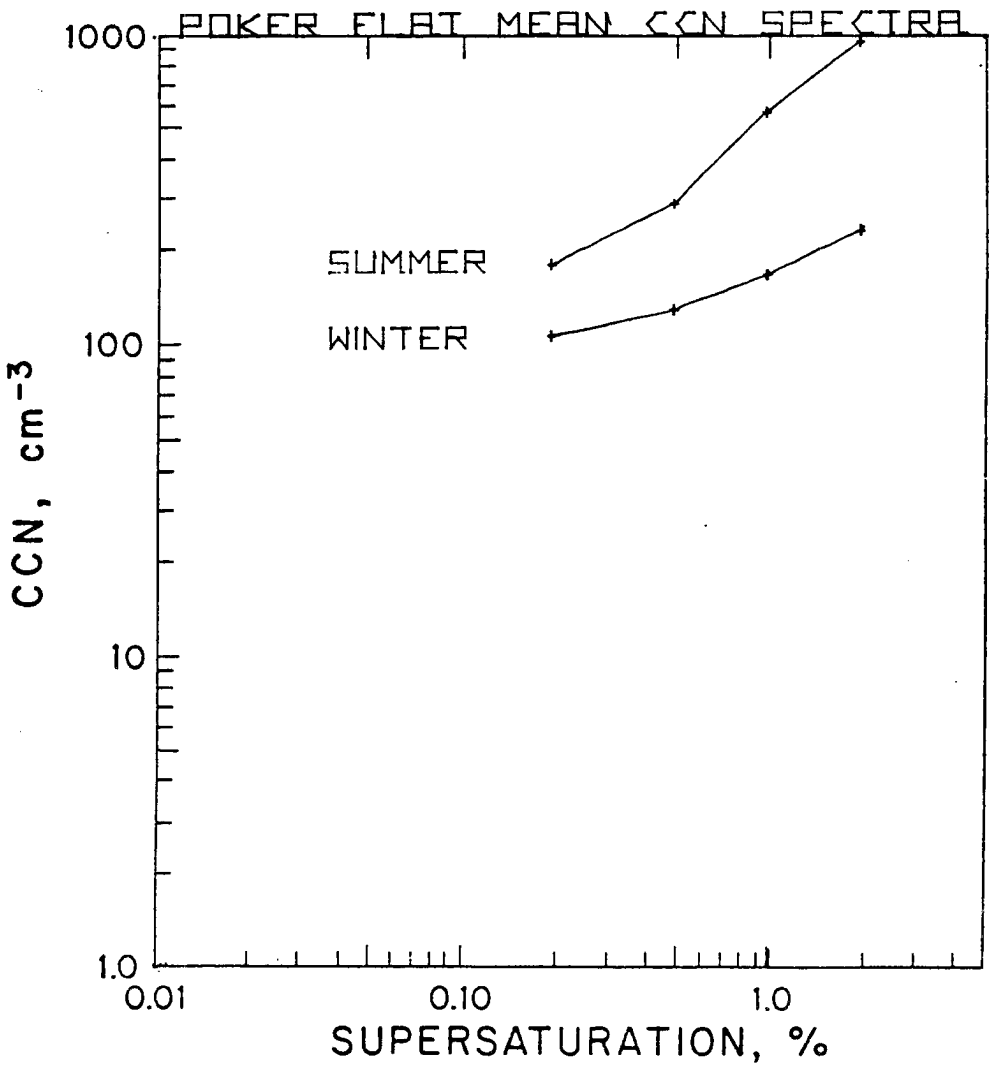


Fig. G.7 Mean CCN spectra for Poker Flat, Alaska, USA, derived from seventeen spectra measured during summer, 17 - 25 July, 1979 and twenty spectra measured during winter, 4 - 13 April, 1980.

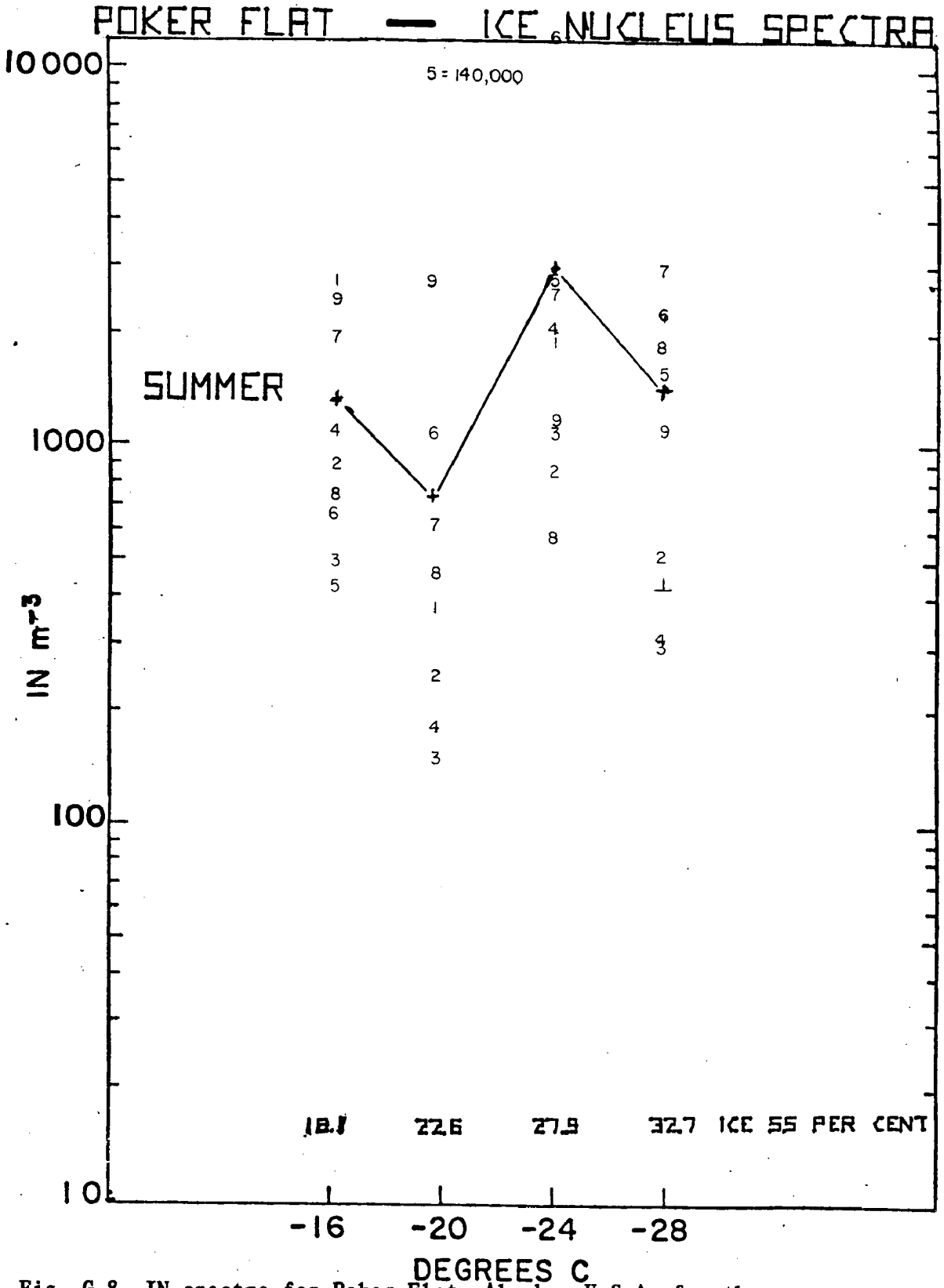


Fig. G.8 IN spectra for Poker Flat, Alaska, U.S.A. for the summer experiment 17-25 July 1979. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate a less-than-blank concentration. The mean spectrum is ploted using only values above blank and does not include sample 5 at -20°C.

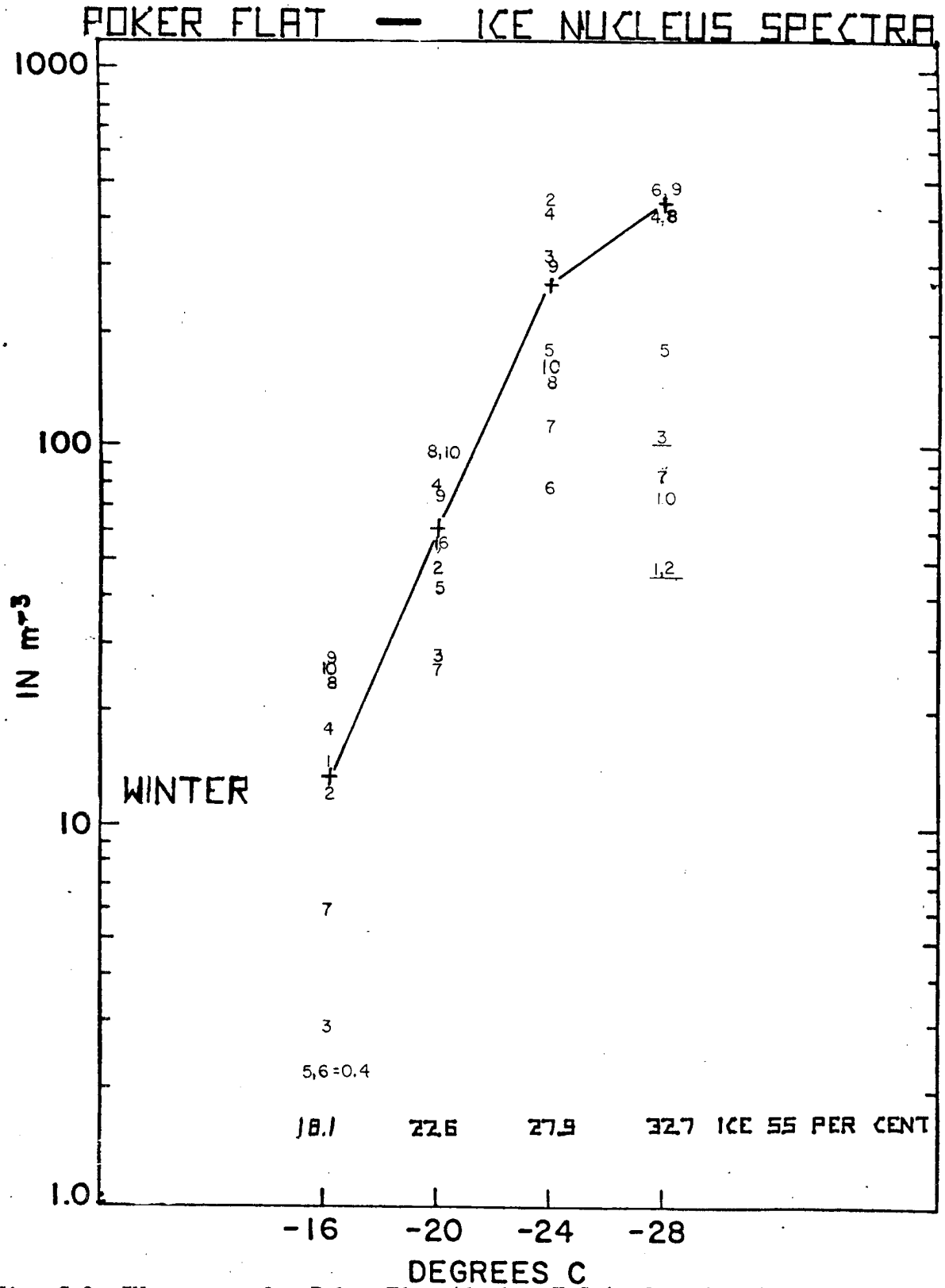


Fig. G.9 IN spectra for Poker Flat Alaska, U.S.A. for the winter experiment 5–14 April 1980. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate a less-than-blank concentration. The mean spectrum is plotted using only values above blank.

aerosol concentrations by season when compared to the high arctic site of Barrow. The data shows that although there is a winter influence of anthropogenic aerosol, other apparently natural sources during summer dominate the cloud active aerosol concentrations. This is true more so for the IN than for the CCN.

G.3a Sources of cloud-active aerosol

The explanation of the observed concentrations and variations of the cloud-active aerosol at Poker Flat as a result of one or two dominant sources is not possible. There were no significant positive correlations between CCN and IN and any of the aerosol source tracers. Regional sources of $SO_4^=$ are apparently important during the winter, but CCN concentrations are higher during the summer when the $SO_4^=$ concentrations are lowest. It is interesting to note that both during summer and winter the highest concentration of I corresponded to the highest concentration of CCN. One possible explanation of higher CCN during the summer versus the winter is the greater amount of biogenic activity during the summer. I is assumed to indicate secondary marine aerosol production and transport to the site. If this is true, it would be expected that the I may be indicative of the proper conditions for the production of secondary aerosol from natural sources.

The IN concentrations are also difficult to interpret. The sudden high concentrations of IN during the middle of the summer sample period cannot be attributed to any other sudden increase in aerosol source tracer components. As previously mentioned, the weather during this sample consisted of light to moderate rain and fog. There have been other reports in the literature of the peaking of IN concentrations as a result of the onset of precipitation. This may be a result of enhanced

vertical mixing and the bringing to the surface of aerosol aloft with a high number concentration of IN. As evidenced by Figures G.3c and d, and the lack of a mass increase in the composition of the aerosol, these aerosol are small (approximately 0.1 μm radius). Their source is unknown.

One of the reasons for choosing this site was to assess the importance of aerosol transport from, and over, the North Pacific to the high-arctic as a source of cloud active aerosol, and the aerosol in general. During the winter, concentrations of all aerosol tracers were less at Poker Flat than at Barrow, except for Al. During the summer, when Barrow aerosol is representative of background conditions, the concentrations of Al, V and $SO_4^=$ are all higher at Poker Flat. For the summer case, transport from the south toward Barrow may account for some of the aerosol trace chemistry at Barrow, but cannot account for the winter situation. Because of this, the observed higher concentrations of CCN and IN at Poker Flat during winter are not related to the CCN or IN at Barrow. There is little to no difference between the two sites in concentration of CCN and IN during summer. Thus, the two sites are effectively separate in terms of their aerosol climatology. Any exchange is probably most important during the winter in a north to south direction, via a path around the eastern end of the Brooks Range, along the Yukon and Porcupine River valleys. Certainly transport via the North Pacific pathway to the high arctic was not important during this study in determining the aerosol trace element composition and the cloud active aerosol concentrations for the high-arctic as represented by the Barrow study site.

The most important result of the cloud active aerosol study at Poker Flat was the association of these aerosols with natural aerosol elemental tracers or the lack of strong evidence of an anthropogenic source. The results are not as clear as those from Barrow where the winter anthropogenic aerosol has a dominant effect on the CCN and IN concentrations.

APPENDIX H

DETAILED FIELD EXPERIMENT DISCUSSION
Pangnirtung, NWT, Canada

This site was chosen to lie along a potential transport path from the northeastern USA to the Arctic to the west of Greenland. The location is not a high arctic site. It is located near the Arctic Circle, 2200 km north of Nova Scotia. It is a maritime site since it is located on the southeast coast of Baffin Island.

H.1 Summer Study

The summer aerosol experiment was conducted during the period 22 June to July 1, 1979. Nine, 24-hour sample sets were collected and analyzed. The results of the bulk aerosol trace element chemistry is presented in Table 7, in Appendix A. The summary plot of the aerosol elemental tracers is given in Figure H.1. Figures H.2a-d are the isohypse analyses for the 850 mb and 700 mb pressure levels. SEM photomicrographs are presented in Figure H.3 of aerosol-particles collected from samples 5 and 9.

As has been the case at the other arctic sites during summer, the aerosol loading is low and the pollutant tracer, noncrustal V, was not detectable. Unfortunately, it was not possible to obtain nonmarine sulphate analyses for these samples. However, the results would probably be similar to those of the V. This site is located in a region of mountains and tundra. Vegetation is at a minimum here in comparison to Poker Flat, where the site was located in a forested region. The 850

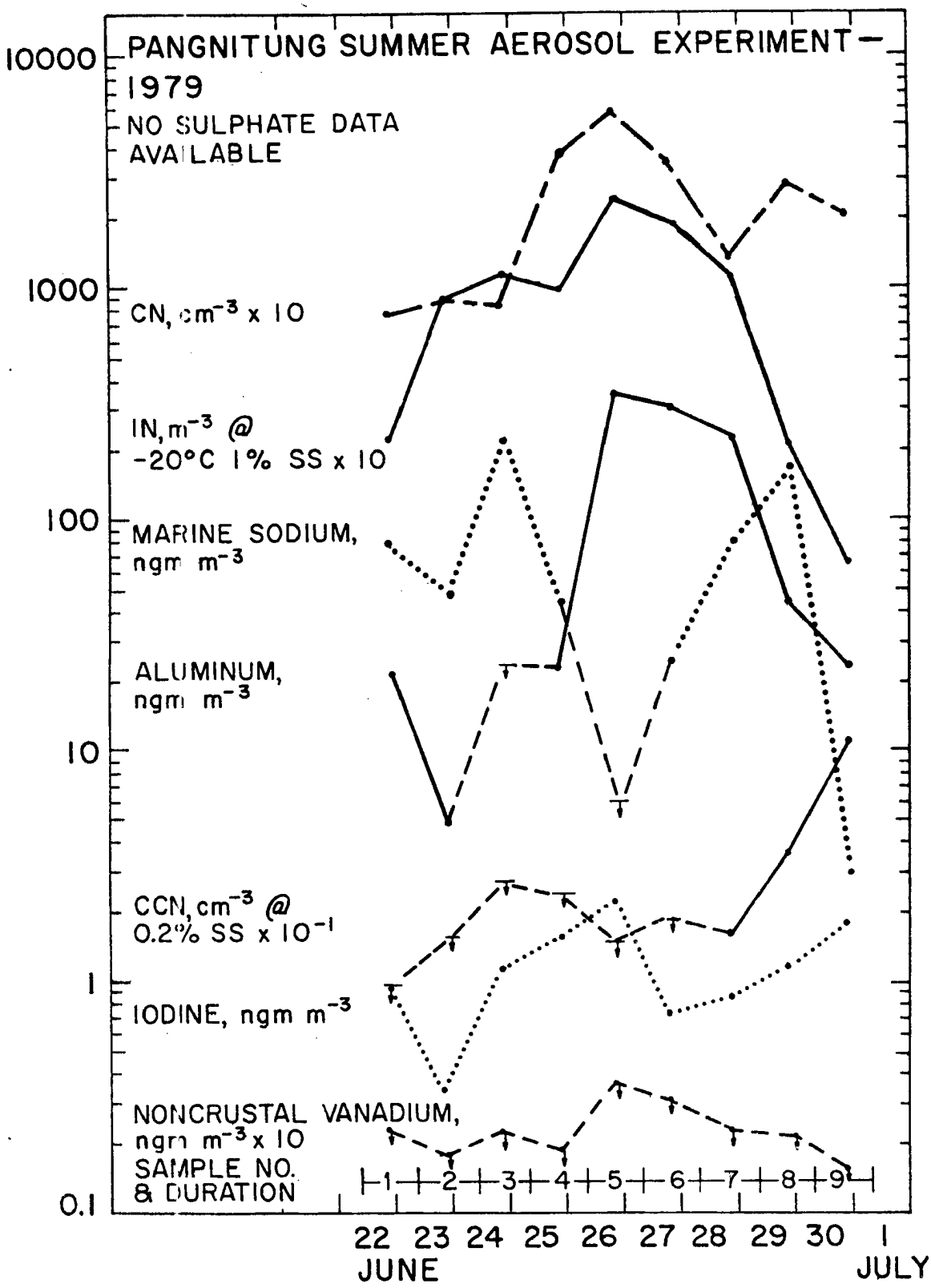


Fig. H.1 Pangnirtung, NWT, Canada summer aerosol experiment, June - July 1979.

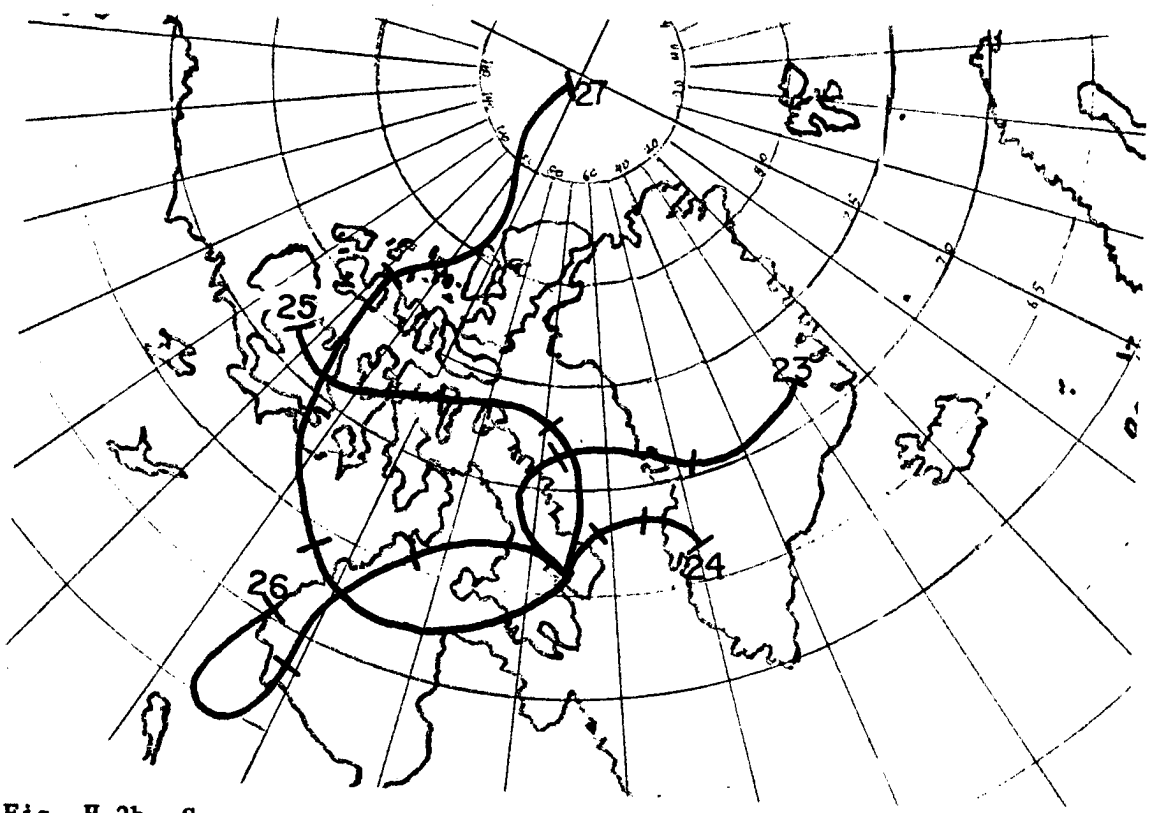


Fig. H.2b Same as a at 700 mb.

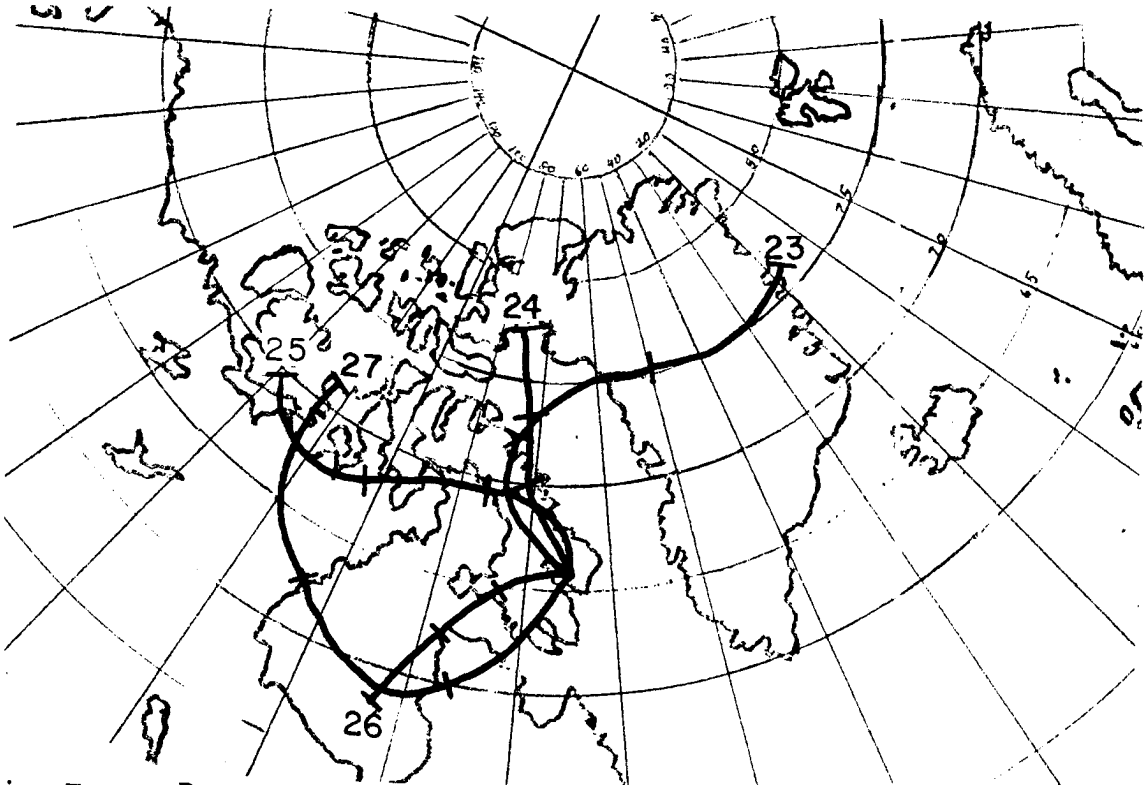


Fig. H.2a Pangnirtung 850 mb isohypse analyses, 23 - 27 June, 1979, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

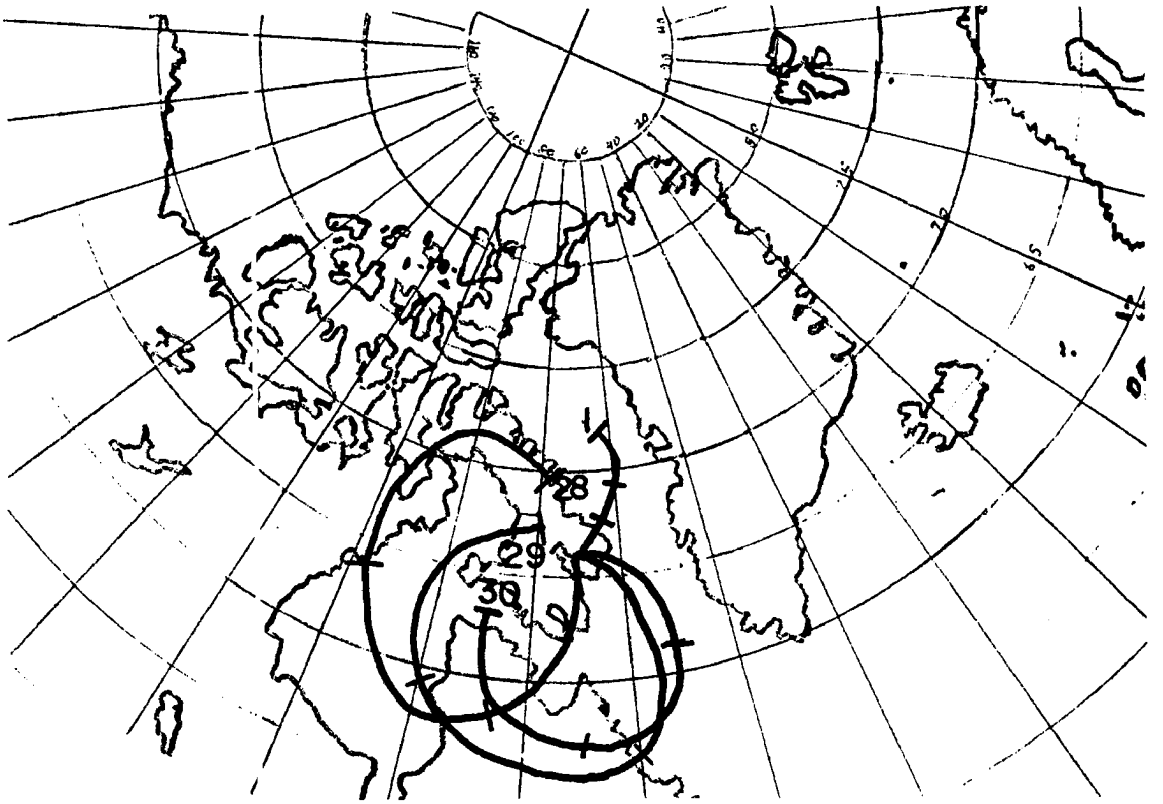


Fig. H.2d Same as a at 700 mb, 28 - 30 June and 1 July, 1979.

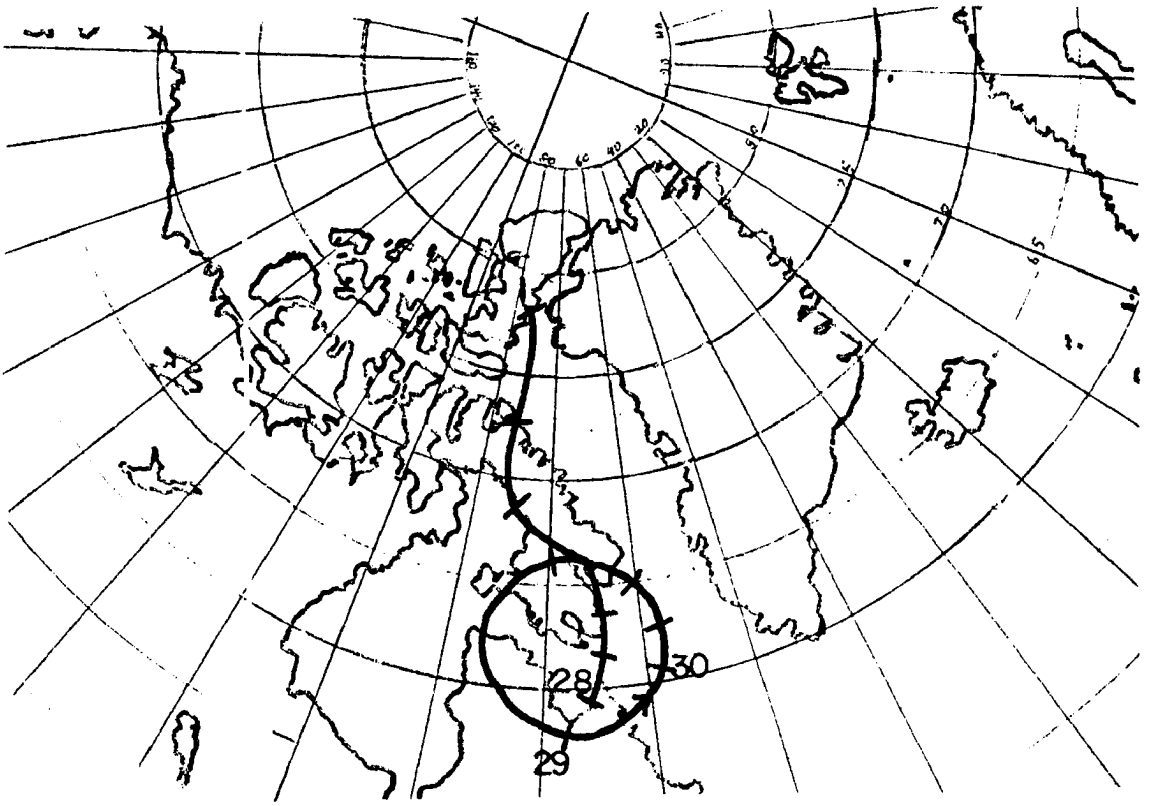


Fig. H.2c Same as a at 850 mb, 28 - 30 June and 1 July, 1979.

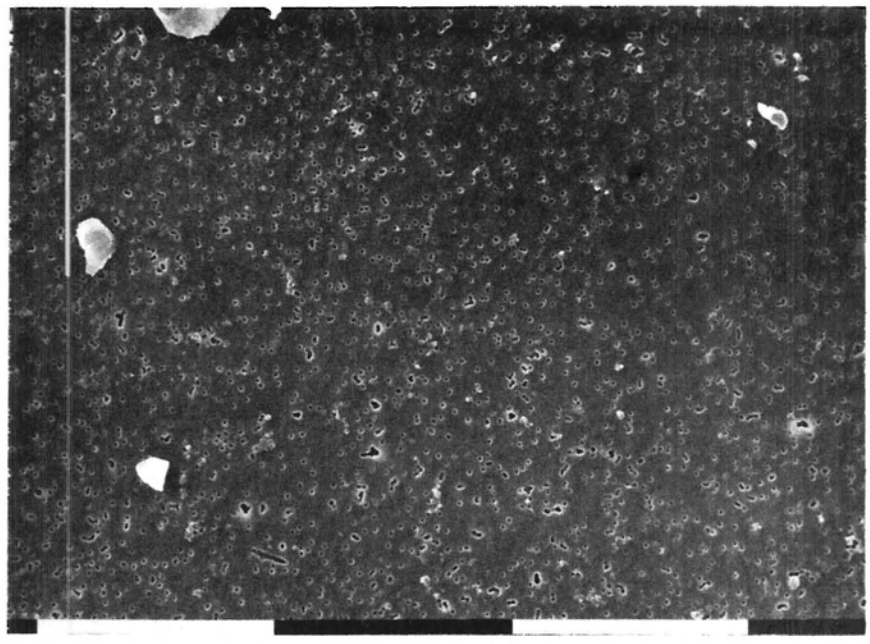


Fig. H.3a Scanning electron microscope photographs of aerosol particles collected at Pangnirtung during summer on 0.2 μm diameter pore size Nuclepore filters from sample #5, 3100X magnification, bar scale is 10 μm.

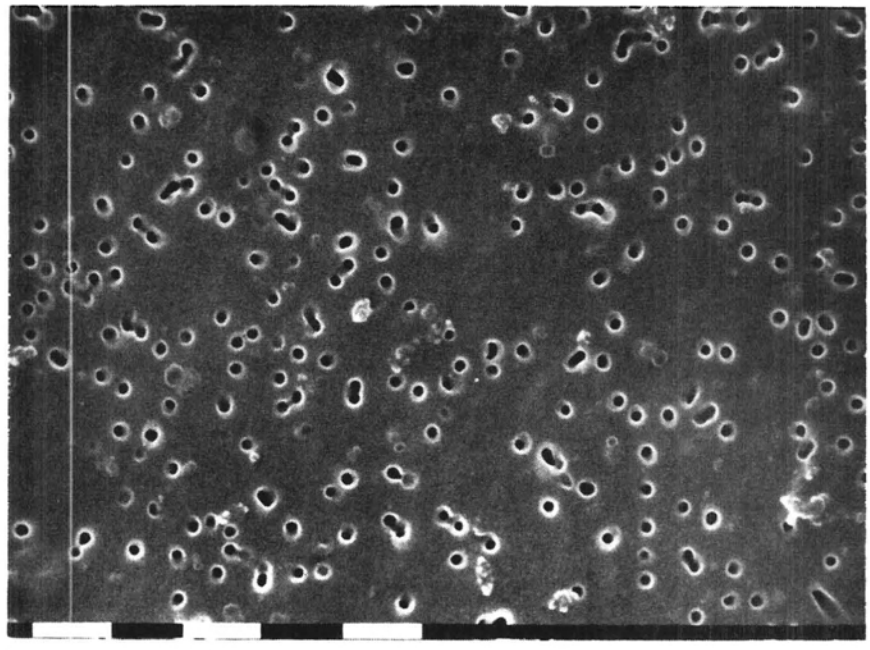


Fig. H.3b Same as a but 10,000X magnification, bar scale is 1 μm.

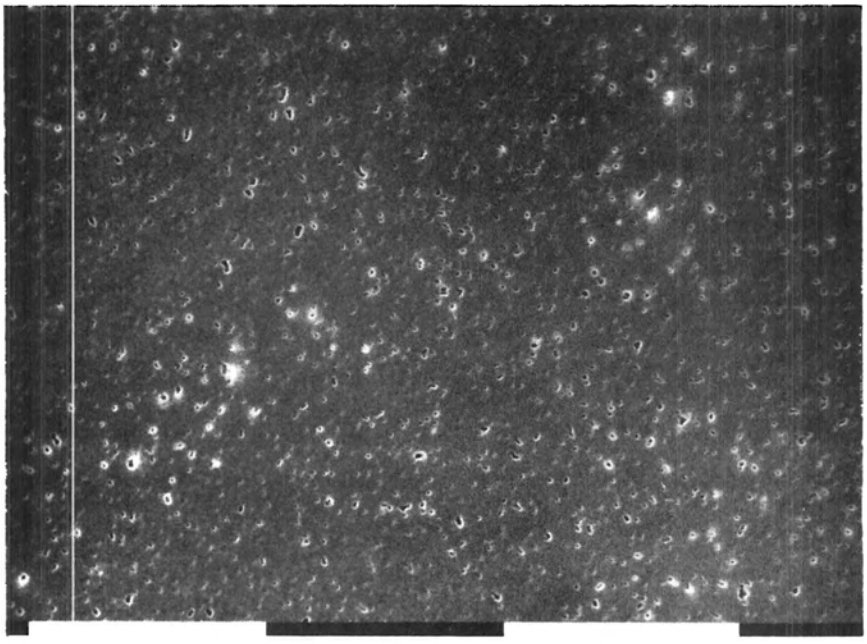


Fig. H.3c Same as a but for sample #9.

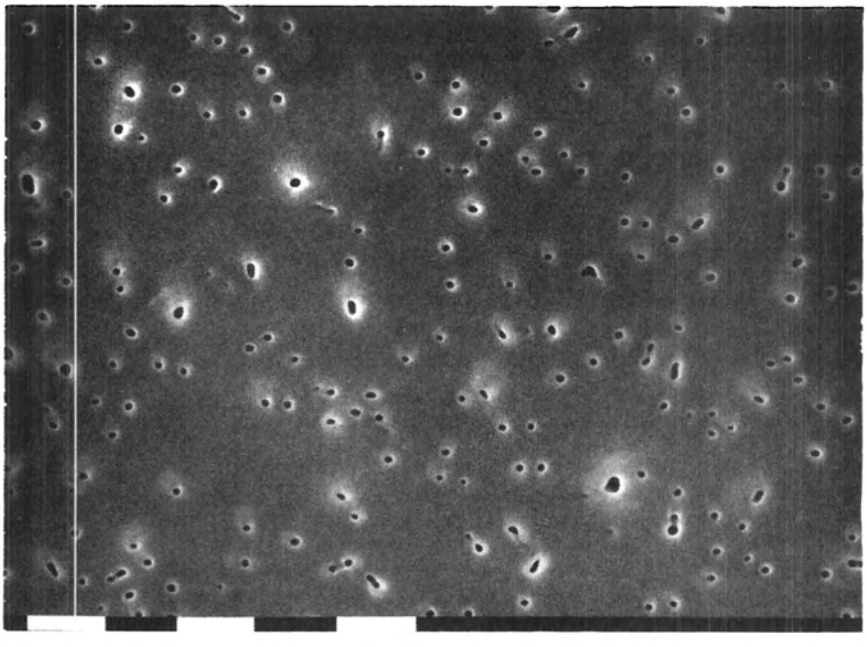


Fig. H.3d Same as c, but 10,000X magnification, bar scale is 1 μm.

mb isohypse analysis shows the winds to be primarily from the southeast to northwest through west. Much of the period was influenced by a low pressure center to the south of the site.

Because of the location of the site within a fjord with a village nearby, local contamination was a possibility on many samples. Sample 5 was collected during the influence of local emissions. During the summer, local emissions were dominated by auto and aircraft exhaust and dust from the local roads and airfield. Some light heating oil was also burned. The influence of sample 5 by local emissions is evident from the CN concentrations which peaked during this sample. As a result, all other aerosol tracers also peaked except for marine Na which had one of the lowest concentrations of the period. This sample was characterized by local downslope winds which carried emissions from the village to the sampling site. Later, for samples 8 and 9, the synoptic situation brought flow from around the leading edge of a low pressure system to the site from the southeast.

H.1a Cloud condensation nuclei

The CCN concentrations measured at 0.2% SS at this site are lower than either those measured at Poker Flat or Barrow. Samples 1-6 had concentrations below the detection limit of about 15 cm^{-3}. The concentrations rose rapidly during samples 7, 8 and 9. These were characterized by flow from the southeast, with no local influence. Referring to Table 5.2, the CCN were not correlated significantly with any of the aerosol tracers. The greatest positive correlation occurred with I. This suggests, along with the isohypse analyses, that there are greater numbers of CCN associated with airmasses transported from the southeast. CCN concentrations increased from less than 20 cm^{-3} to

greater than 100 cm^{-3}. The lack of correlation with measured aerosol tracer components suggests a relationship with an unmeasured species. Based upon sample 5, there is positive evidence that local emissions of pollutants are not sources of CCN.

H.1b Ice nuclei

The variation of IN at this site during summer can be readily attributed to the variation of Al and CN. Referring to Table 5.2 it is evident that IN are positively correlated to the Al concentrations and less importantly to the CN concentrations. The highest IN concentrations correspond to the highest Al and CN concentrations. This suggests local road dust associated with vehicular traffic (resulting in the high CN concentrations) is responsible for the high IN concentrations. Figure H.3 shows the contrast of samples 5 and 9 in the number of dust particles present. Virtually no aerosol particles are seen on the sample number 9 filter. Numerous bright particles are evident on the filter from sample 5. These bright particles are typically dust particles and can be related to the maxima in Al concentrations as shown in Figure H.1. Because of this influence of local activity, it is impossible to assess the importance of other potential sources.

H.2 Winter Study

The winter aerosol experiment was conducted during the period 26 January to 4 February, 1980. Nine samples were collected and analyzed. The samples were not collected over continuous 24 hour periods. Rather, the pumps used for the aerosol collections were controlled by wind direction and speed. Whenever the wind speed was above the threshold velocity of the wind sensor (approximately 2 m/sec) or the wind

direction was certain to exclude air which had passed over the village of Pangnirtung, the pump was switched on. The total sample time was restricted to 24 hours. However, a few of the samples took up to two days to complete. The results of the bulk aerosol trace element chemistry is presented in Table 8, Appendix A. The summary plot of the aerosol elemental tracers is given in Figure H.4. Figures H.5a-d are the isohypse analyses for the 850 mb and 700 mb pressure levels. SEM photomicrographs are presented in Figure H.6 of the aerosol-particles collected from samples 3 and 5.

As evidenced by Figure H.5, the circulation during the winter was, in general, stronger and dominated by flow from the southeast. Flow from the northwest occurred during the last sample collected. The dominant southeast flow is a reslt of the semipermanent position of the Icelandic low located to the south and east of this site during the inter. This low pressure system is usually well developed and has strong wnds associated with it. Pangnirtung is located on the northwest side of this low most of the time during the winter season. Winter storms form near Cape Hatteras, move up the east coast of the USA, and intensify. They then move to the east-northeast and become associated with the Icelandic trough. It is this pattern of storm movement and upper winds which are conducive to transport to the north, west of Greenland. Therefore, the winter period provides the best conditions for assessment of this transport path.

In general, the aerosol particle tracer components plotted in Figure H.4 show a large degree of variability. Concentrations of all of the components are slightly higher than those of the summer experiment. Marine Na, I, Al and CN all are between 1-2 times higher in

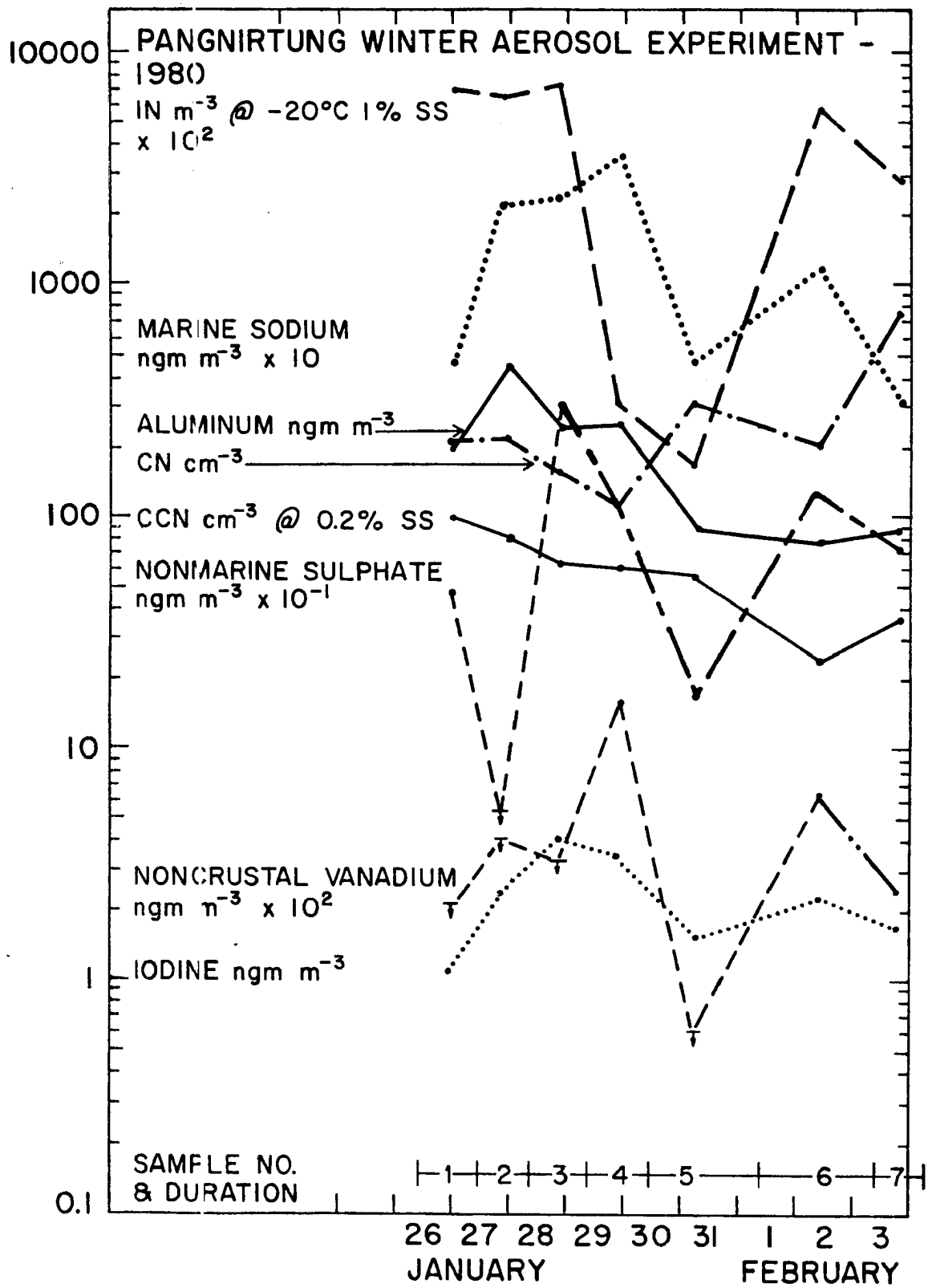


Fig. H.4 Pangnirtung, NWT, Canada winter aerosol experiment, January - February 1980.

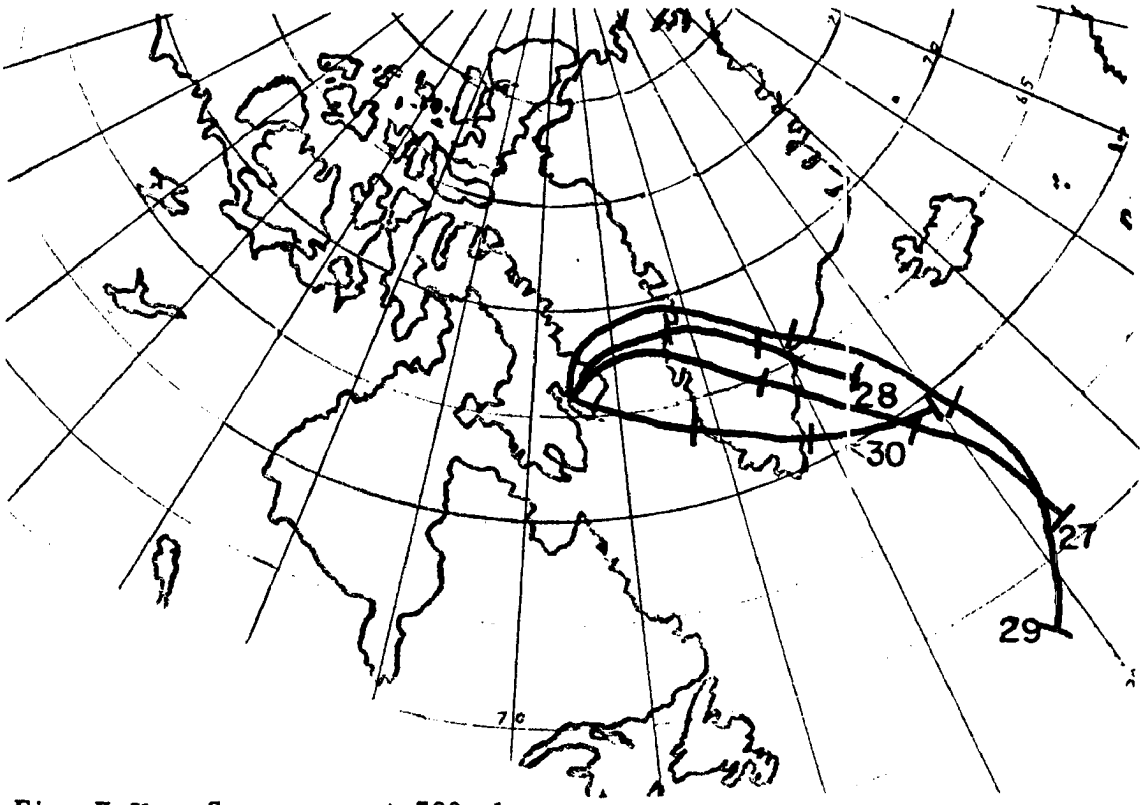


Fig. H.5b Same as a at 700 mb.

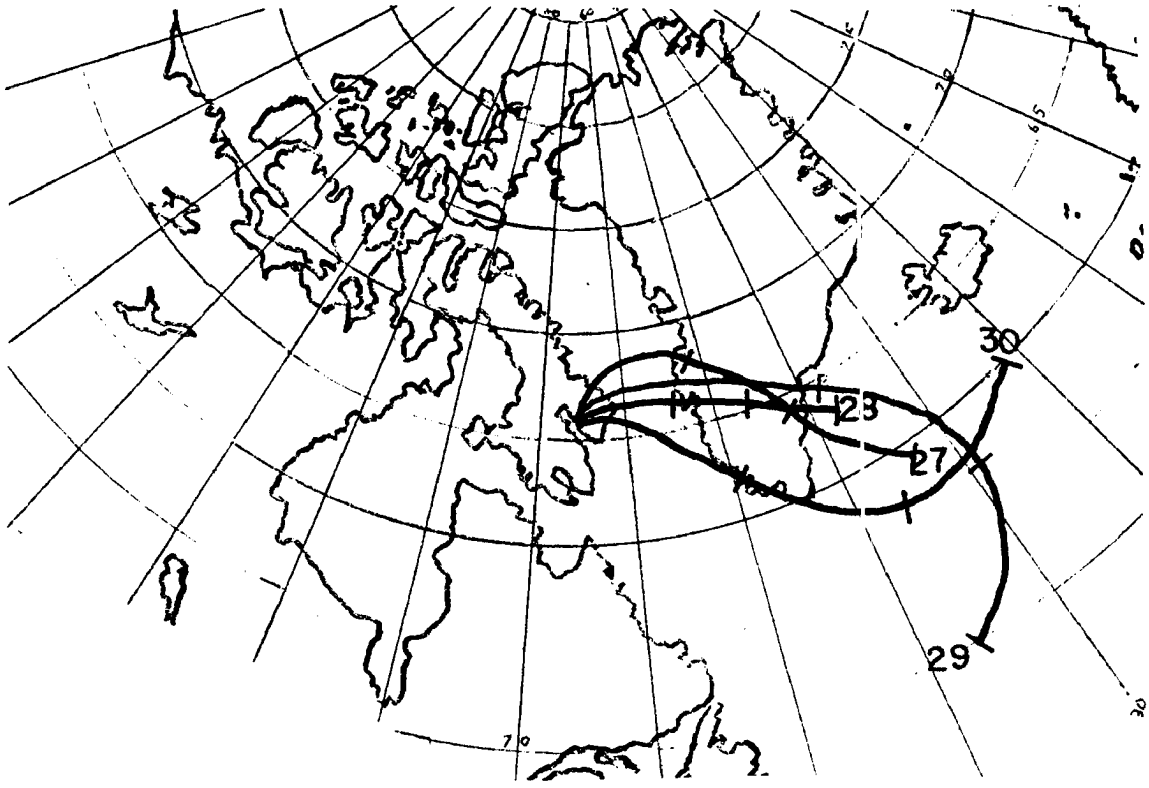


Fig. H.5a Pangnirtung 850 mb isohypse analyses, 27 - 30 January, 1980, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

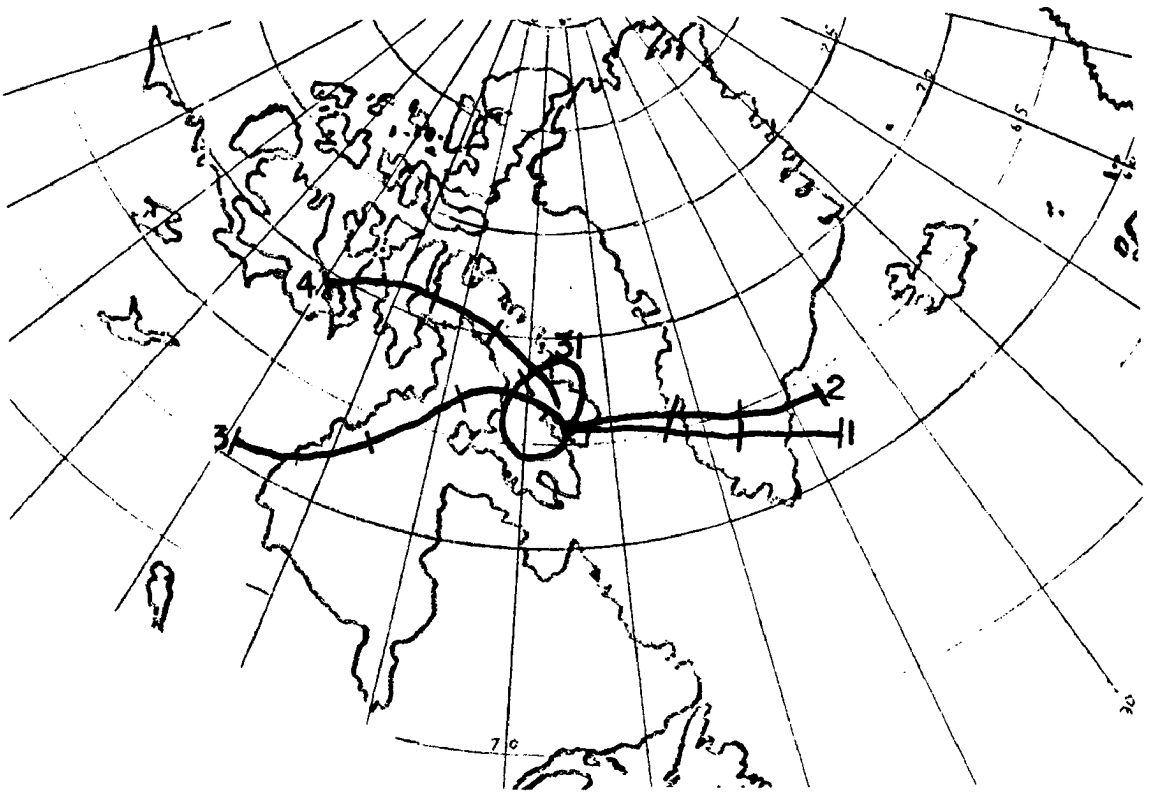


Fig. H.5d Same as a at 700 mb, 31 January - 4 February, 1980.

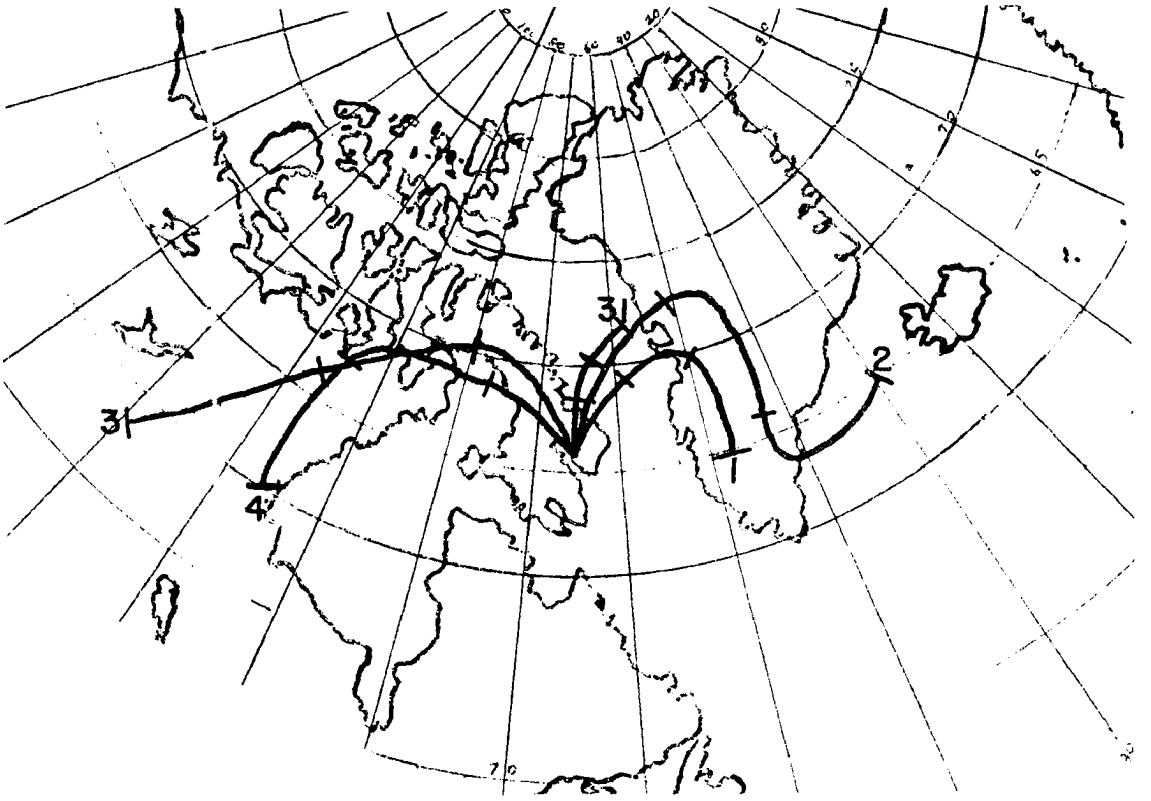


Fig. H.5c Same as a at 850 mb, 31 January - 4 February, 1980.

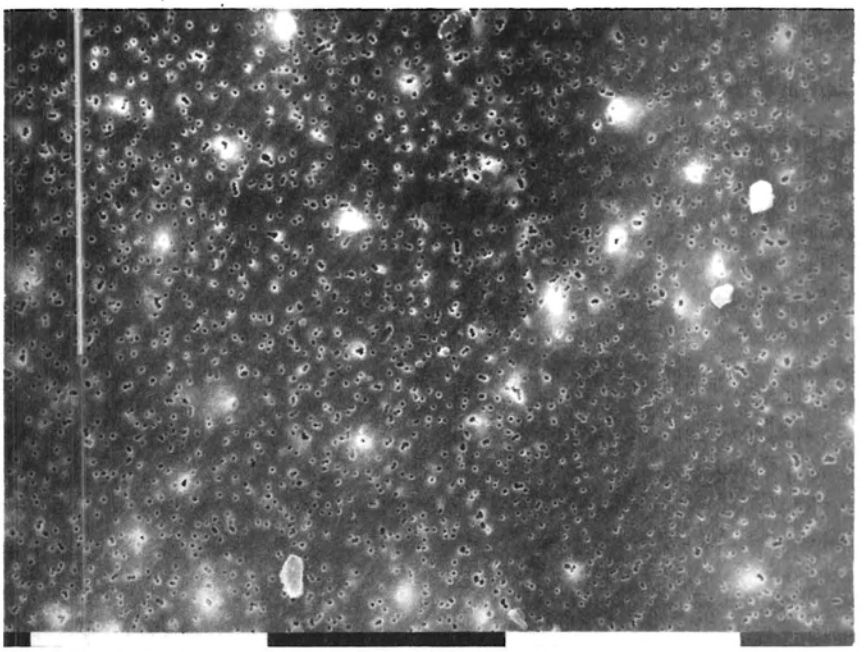


Fig. H.6a Scanning electron microscope photographs of aerosol particles collected at Pangnirturng during winter on 0.2 μm diameter pore size Nuclepore filters from sample #3, 3100X magnification, bar scale is 10 μm.

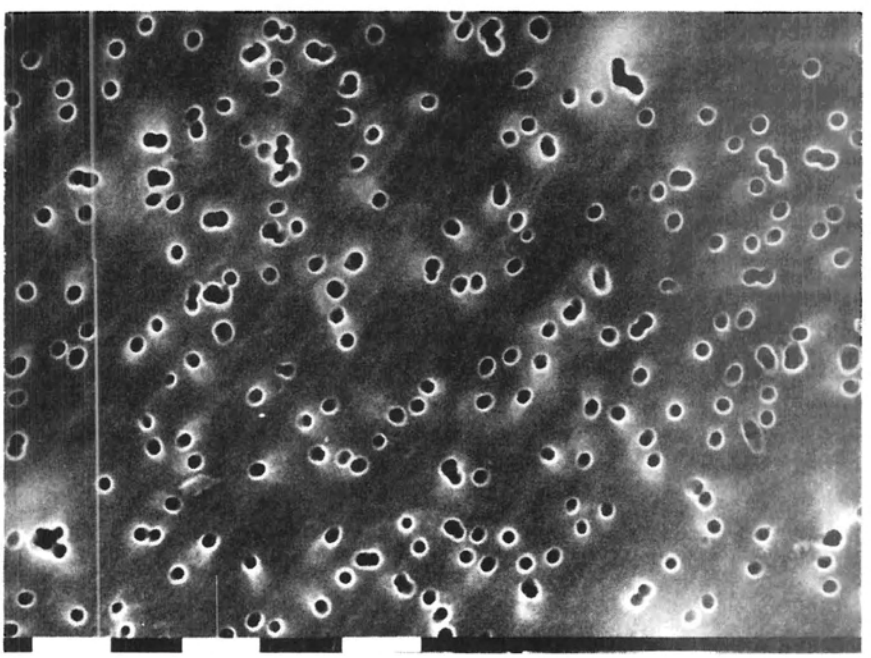


Fig. H.6b Same as a but 10,000X magnification, bar scale is 1 μm.

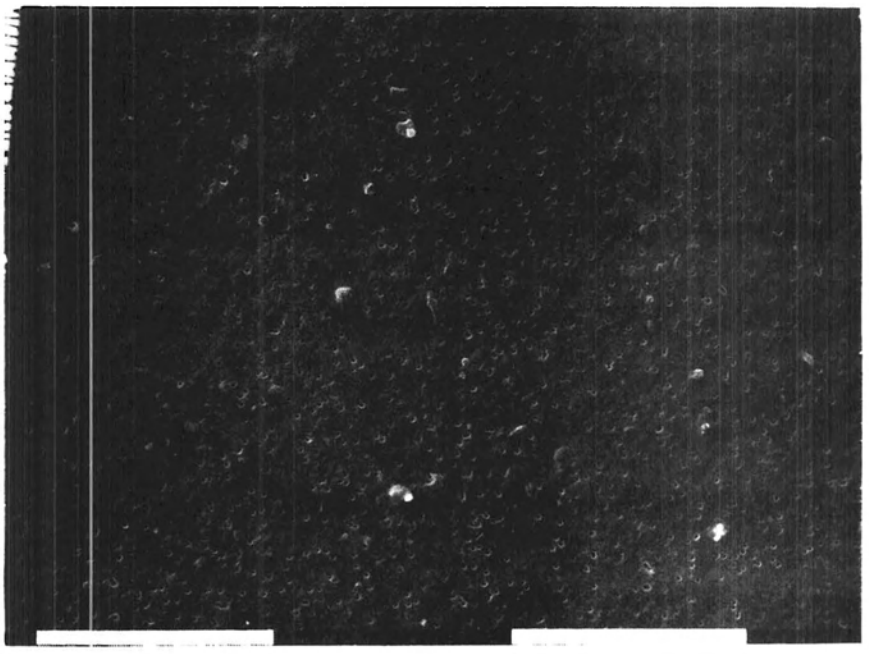


Fig. H.6c Same as a but for sample #5.

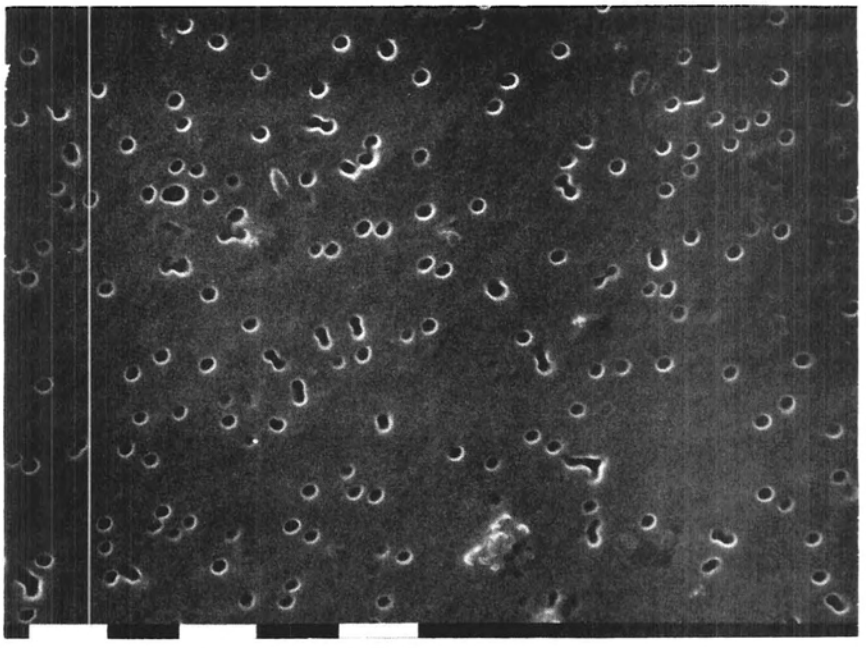


Fig. H.6d Same as c, but 10,000X magnification, bar scale is 1 μm.

concentration than they were during the summer. Noncrustal V has the highest increase of a factor of 7.7. No comparison is possible for $SO_4^=$ due to the lack of summer data. However, both of these pollutant aerosol tracers were not detected in some samples. Thus, although they may be present in higher concentrations, they are still near their detection limits. These concentrations are 3 to 20 times lower for $SO_4^=$ and V, respectively, than the concentrations at Barrow during winter.

H.2a Cloud condensation nuclei

The CCN concentrations show a slow decrease with time during the sampling period as shown in Figure H.4. The mean concentrations are about twice those measured during the summer experiment. Referring to Table 5.2, no significant positive correlation occurred between CCN and any of the aerosol elemental tracer components. Figure H.4 shows two distinct periods of marine aerosol maxima occurred during the experiment. The CCN plot shows no dependence on the presence of these aerosol particles, even though their concentrations were high enough to measure accurately. Al, the natural continental aerosol indicator, parallels the CCN the closest. Its occurrance at concentrations higher than any other arctic site during winter suggests a local source of dust. The SEM photomicrographs of aerosol particles from samples 3 and 5 show the presence of dust particles. Because of the few data points, little can be said of the relationship of the CCN to the pollutant tracers, as Table 5.2 indicates. The CCN concentrations at Pangnirtung during winter are the lowest of any arctic site studied including those to be described in later appendices. This is probably a result of airmasses arriving at Pangnirtung from the southeast, from mid-latitudes, but after having been associated with the Icelandic low, one

of the stormiest regions in the Northern Hemisphere. Removal processes associated with this stormy region effectively filter out any aerosol, especially the cloud-active aerosol. This is why pollutant aerosol particles from the northeastern USA, which are advected to the east into these storm systems, have a low probability of reaching Pangnirtung or locations further north.

H.2b Ice nuclei

The IN concentrations during the winter experiment show no relationship to any of the aerosol particle tracers. Only seven samples were collected, making any statistical analyses difficult. The IN do not show a positive correlation with Al as in the summer, although Al concentrations are, in general, higher. Overall, the IN concentrations are lower than were during the summer, and lower than any other arctic site. This is probably attributed to the same removal process mentioned above for the CCN. Poisoning effects due to pollutants cannot be the cause of the low IN concentrations because of the very low pollutant aerosol concentrations. Overall, low aerosol concentrations at this site during the winter, and the small number of samples, do not make it possible to come to any firm conclusion concerning the IN and their possible sources. The result of the summer experiment, local dust sources of IN, may also hold true during the winter. A longer experiment is required.

H.3 Seasonal Differences in Cloud-Active Aerosol

Figures H.7 depicts the seasonal mean CCN spectra for summer and winter at Pangnirtung. Figures H.8 and H.9 depict the individual IN conentrations for summer and winter with the mean spectra also plotted. In addition, Tables 5.3 and 5.4 present the winter/summer ratios of CCN

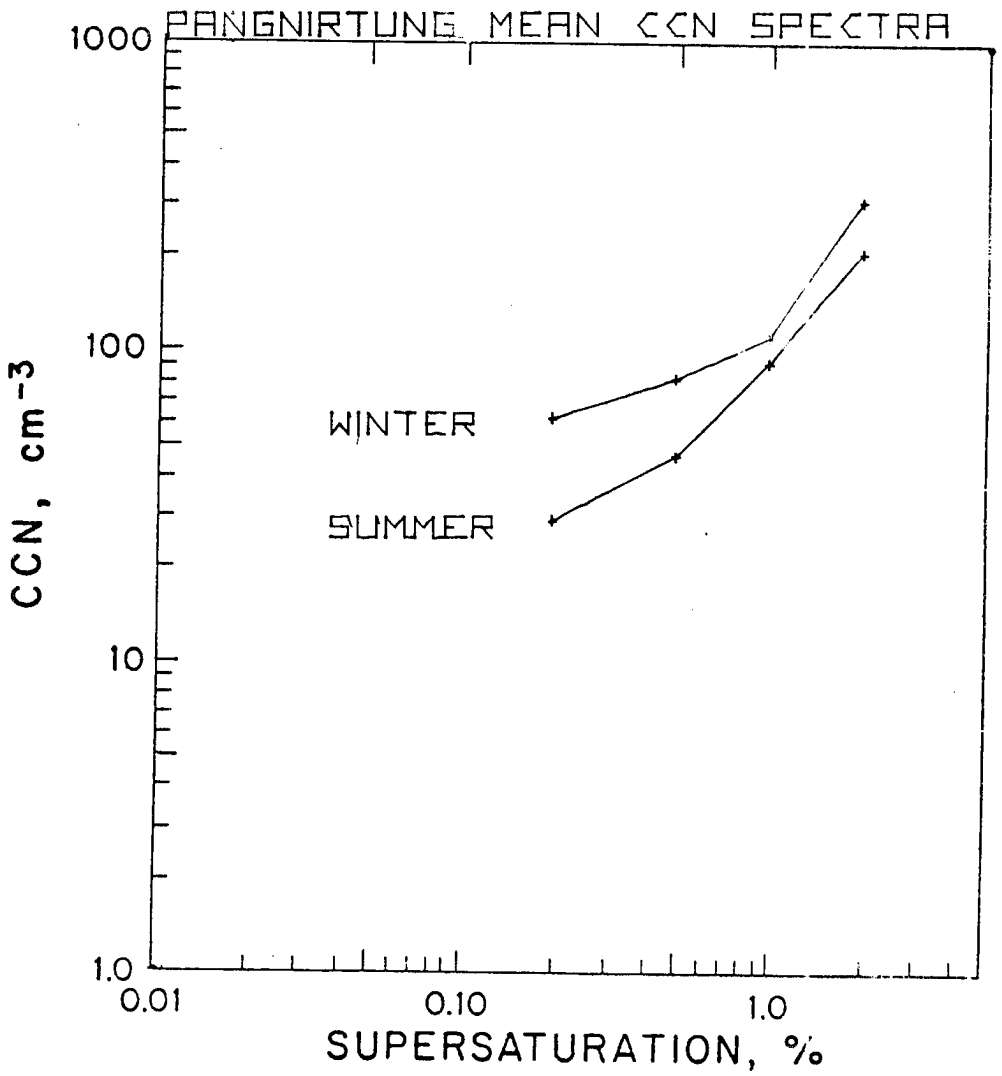


Fig. H.7 Mean CCN spectra for Pangnirtung, NWT, Canada, derived from twenty spectra measured during summer, 21 June 1979 and sixteen spectra measured during winter, 26 January - 3 February, 1980.

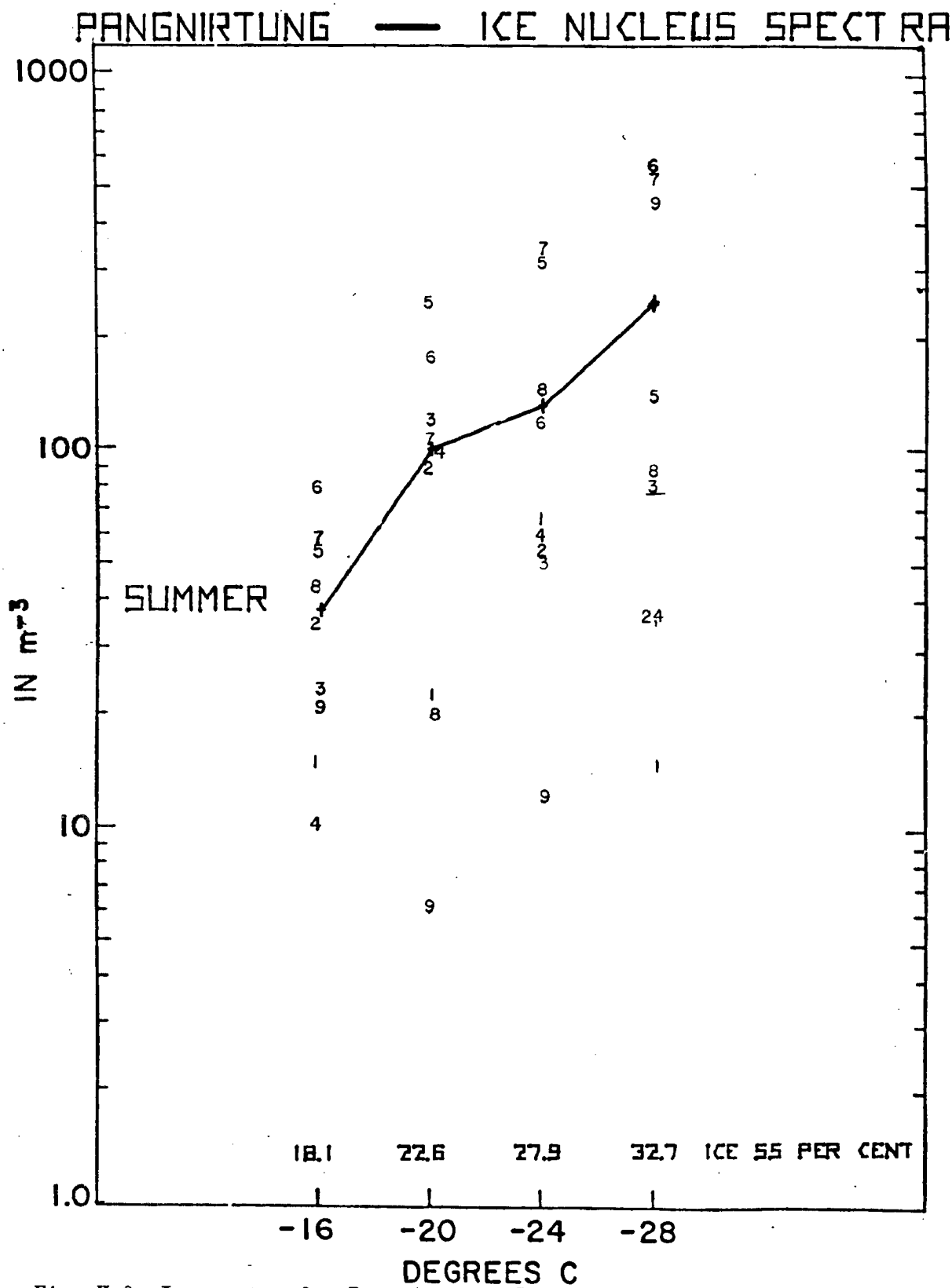


Fig. H.8 In spectra for Pangnirtung, Canada for the summer experiment, 22 June - 1 July 1979. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

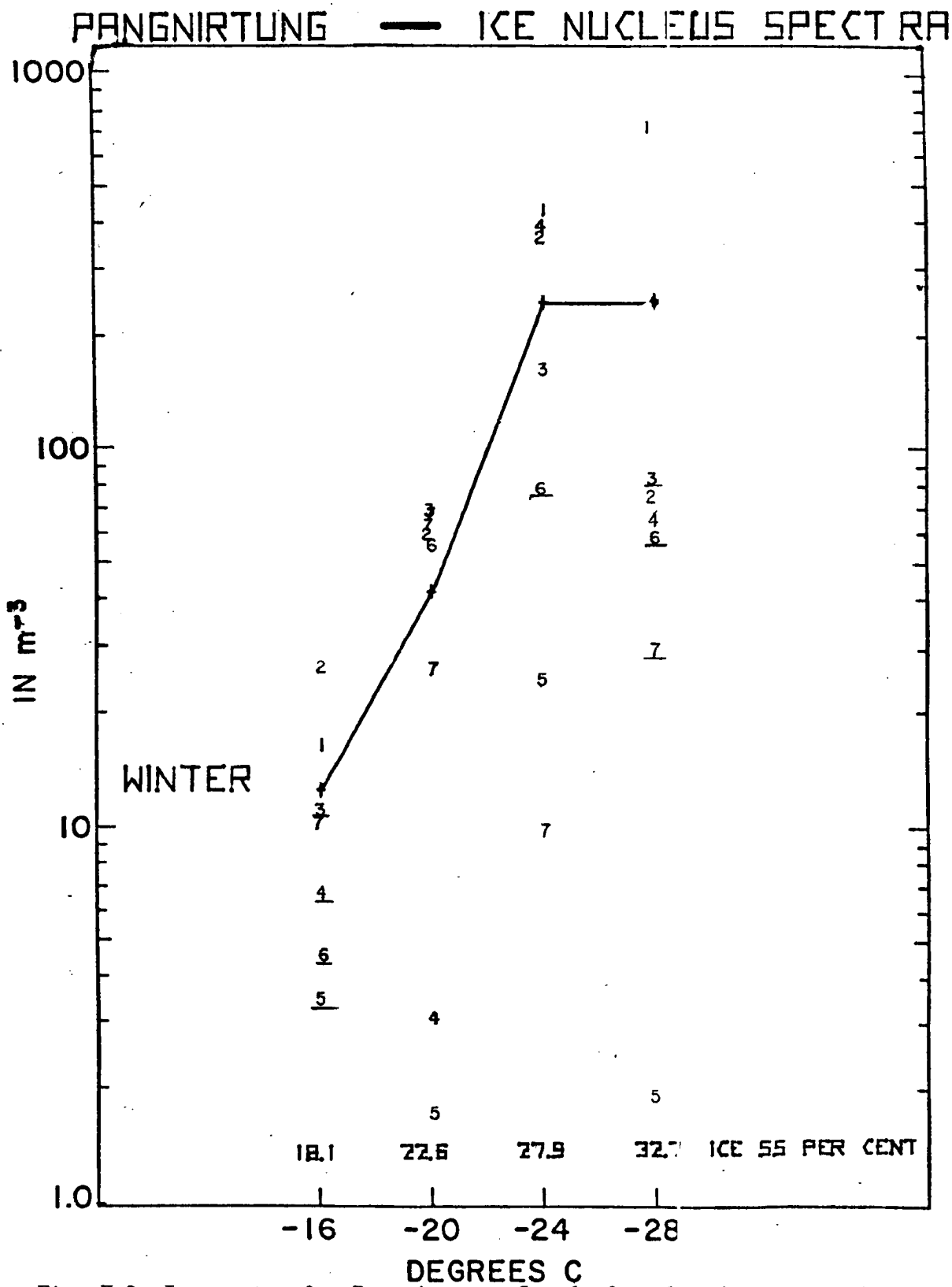


Fig. H.9 In spectra for Pangnirtung, Canada for the winter experiment, 26 January - 3 February 1980. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

at 0.2% SS and IN at $-20^{o}C$ and 1% WSS along with the pollutant aerosol tracer ratios. There is a suggestion in the data that the same factors which influenced the IN and CCN seasonal differences in concentration at Barrow may apply here. That is, the elevated winter pollutants result in higher CCN and lower IN. However, the factor of 7.7 increase in non-crustal V of winter versus summer is a result of a small absolute increase in V imposed upon a very low total concentration. This cannot be used to account for the observed differences in CCN and IN. In general, Pangnirtung is a site typifying a remote "background" location in terms of aerosol concentrations. This is true both summer and winter. Any seasonal contrast of the aerosol is a result of very small perturbations on a low mean aerosol baseline.

Although Pangnirtung is closest of all the arctic study sites to the North American source region, and transport is conducive for advection of aerosol to Pangnirtung from the northeast USA, removal processes along this stormy pathway prove to be more important. From these results, Pangnirtung is located in what can be considered as one of the most "remote" locations from effects of long-range-transport of anthropogenic aerosol of the arctic sites studied. It is in a position that isolates it physically and meteorologically from direct transport from source regions such as Europe and North America and from the aged anthropogenic aerosol of the Arctic to the north in the winter. This conclusion is based on a small set of data and may not apply at other times of the year.

These data fit the pattern observed by Barrie et al. (1981) who measured non-marine $SO_4^{=}$ and non-crustal V at two other Canadian arctic sites at the same time the study was underway in Pangnirtung during the

winter of 1980. The two sites, Mould Bay (119° W, 76° N) and Igloolik (82° W, 69° N) are basically located on a line to the northwest of Pangnirtung. Their results show the two pollutant aerosol tracers to increase in concentration with increasing distance to the north from Pangnirtung. The values of the concentrations of $SO_4^=$ and V at Pangnirtung were 0.5 and 0.1, respectively of the values at Igloolik at the same time. The differences were even greater when compared to the data from Mould Bay. This brings the study at Pangnirtung into perspective.

H.4 Sources of Cloud Active Aerosol

In summary, pollutant sources of IN and CCN advected to Pangnirtung are not important. Although the CCN concentrations are higher during the winter along with the pollutant aerosol tracer noncrustal V, the extremely low pollutant aerosol concentrations indicate their transport to this location is not important. CCN showed no relationship with any of the measured natural aerosol tracers. Their concentration variability may be related to an unmeasured component or to a combination of components which was not discernible from the data. The low concentrations and lack of evidence of a pollutant source suggest the CCN were of natural origin during this study both seasons.

During summer, IN concentrations could be explained by local dust emissions from human activity. During winter, a similar relationship not was found. The lower concentrations of IN during the winter may have been more representative of the regional values, since more care was taken to exclude local sources. The evidence from both winter and summer is that long range pollutant transport to this site is not important and therefore does not affect the IN concentrations. Maritime

aerosol sources are also not linked to the IN concentrations. The measured IN concentrations are therefore representative of background values for this area. As a result, atmospheric transport along a pathway from Northeastern USA via Pangnirtung to the north is not an important route for cloud-active or pollutant aerosol from anthropogenic activity. This pathway does not explain the observed high aerosol concentrations found in the high arctic, further north, during winter.

APPENDIX I

DETAILED FIELD EXPERIMENT DISCUSSION
Irafoss, Iceland

Lying just south of the Arctic Circle near the mid-Atlantic Ocean, this site was chosen as approximately midway between the North American source region and the high arctic. This site is not a high arctic site. It is a maritime site located on the North Atlantic storm track associated with the Icelandic Low.

I.1 Summer Study

The summer aerosol experiment was conducted during the period of 17-26 August, 1979. Nine, 24-hour sample sets were collected and analyzed. The results of the bulk-aerosol trace-element composition are presented in Table 9 in Appendix A. The summary plot of the aerosol elemental tracers is given in Figure I.1. Figures I.2a-d are SEM photomicrographs of aerosol particles collected during samples 4 and 7. Borys and Rahn (1981) discuss the summer experiment in detail. This report is presented as Appendix C. IN data were not available for the report. The CCN data are therefore the only cloud active aerosol discussed and reference to the summer CCN data in that report will be made in this section. The report includes 850 mb isobaric trajectory analyses which apply to the summer period and a discussion of the meteorology.

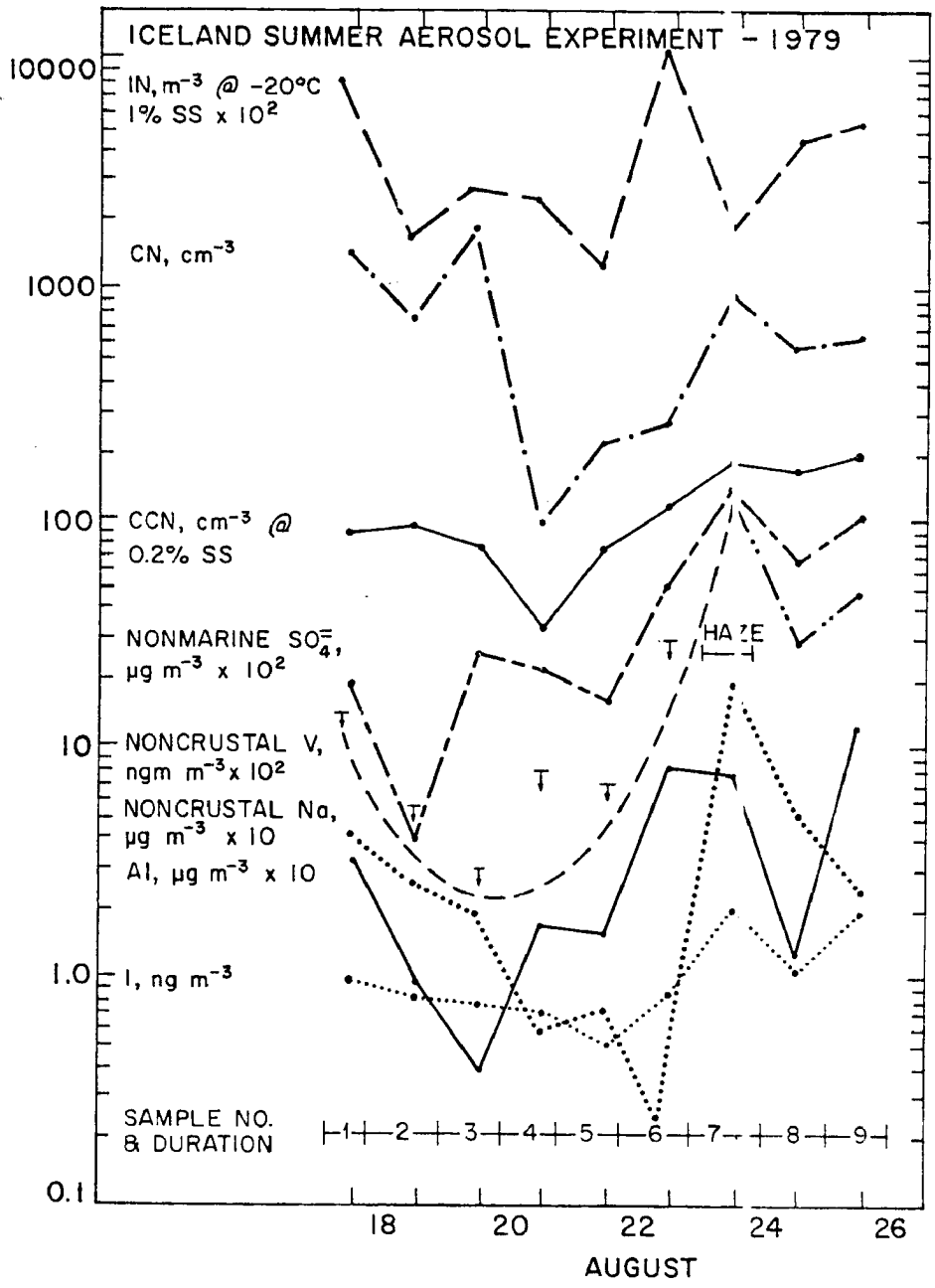


Fig. I.1 Irafoss, Iceland summer aerosol exper ment, August 1979.

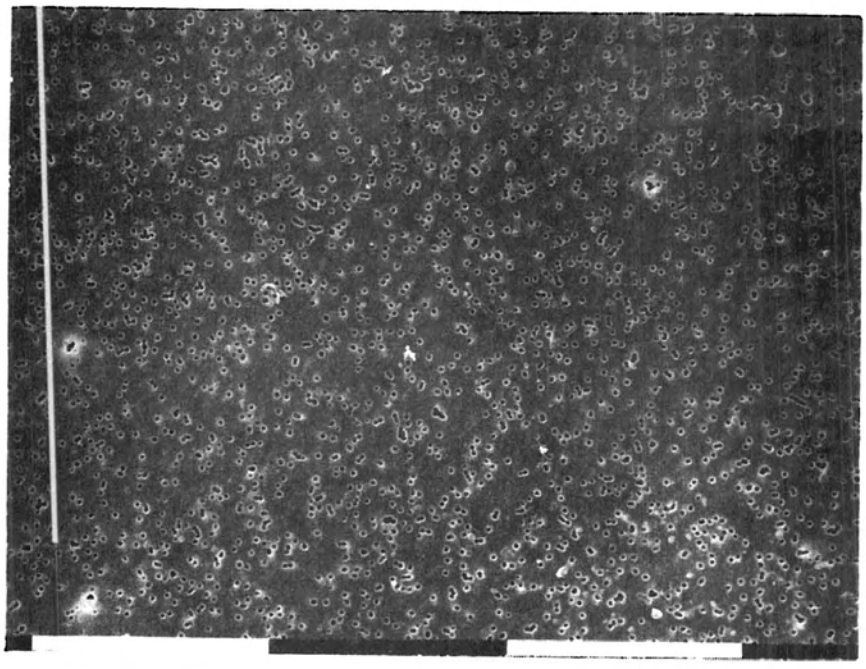


Fig. I.2a Scanning electron microscope photographs of aerosol particles collected at Iraföss during summer on 0.2 μm diameter pore size Nuclepore filters from sample #4, 3100X magnification, bar scale is 10 μm.

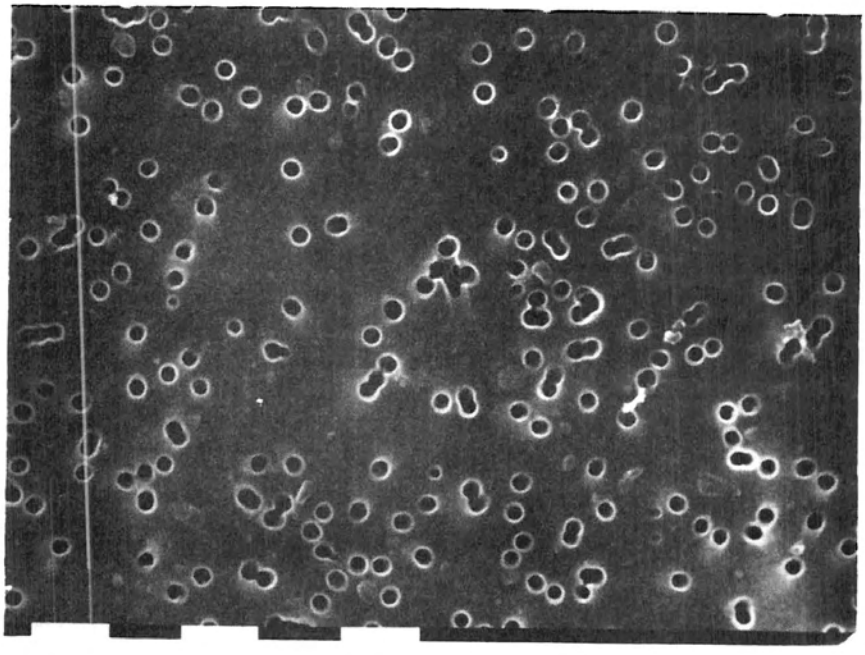


Fig. I.2b Same as a but 10,000X magnification, bar scale is 1 μm.

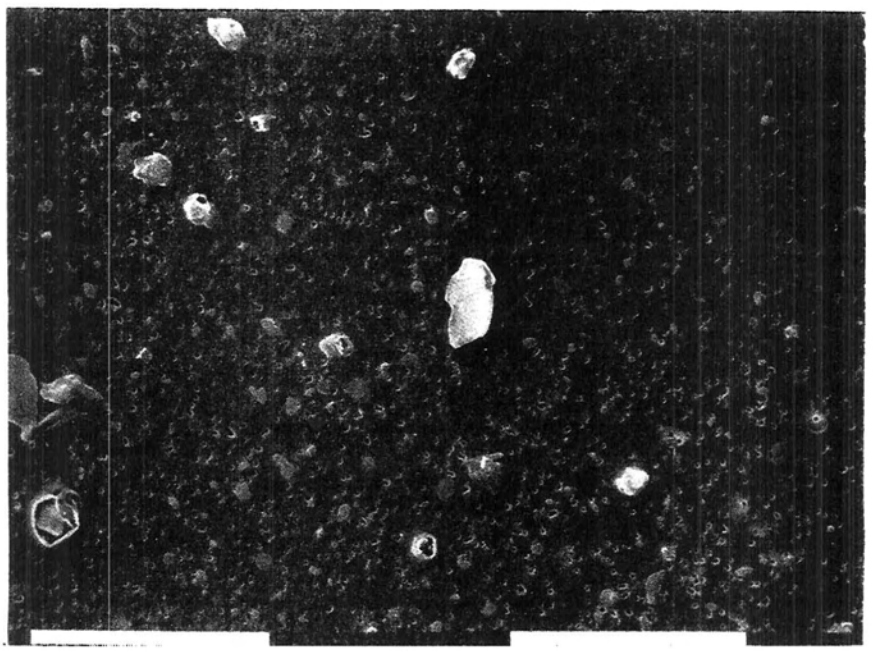


Fig. I.2c Same as a, but for sample #7.

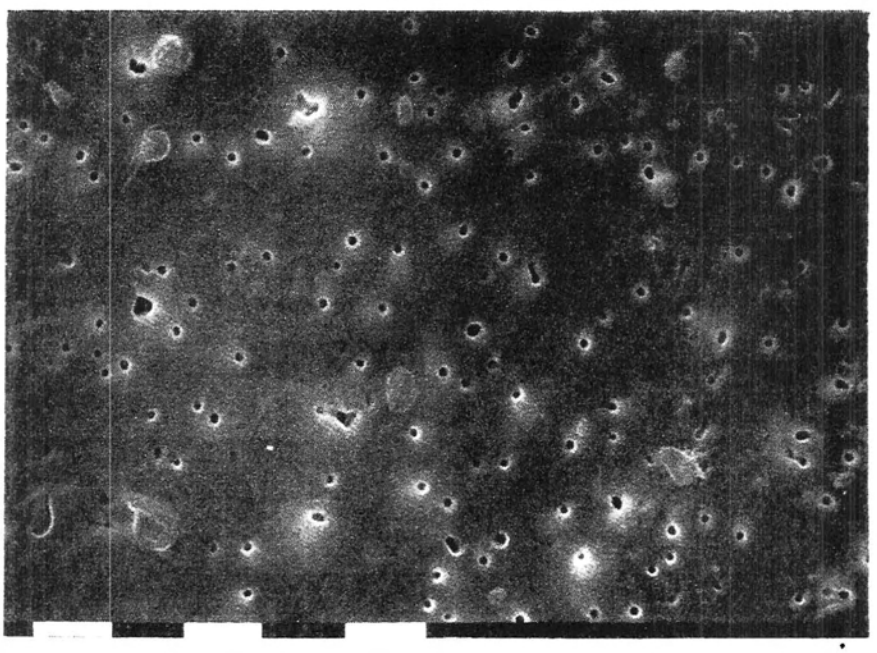


Fig. I.2d Same as c, but 10,000X magnification, bar scale is 1 μm.

I.1a Cloud condensation nuclei

See Appendix C.

I.1b Ice nuclei

The variations of the concentrations of IN at $-20^{o}C$ are presented in Figure I.1. The mean concentration of the IN at this site is the lowest of the arctic sites for the summer by about a factor of two. The transport to the site, as shown by the trajectory analysis in Figure 4 in Appendix C, shows a general veering of the wind from a southwesterly direction through north to northeasterly. The discussion of the aerosol tracers in Appendix C indicates a relatively clean background airmass dominating the samples 1-4 and a gradual influx of an airmass characterized by pollution emissions from Europe, about 2000 km to the east. The change in the aerosol loading from a low in sample 4 to the high in sample 7 can be seen in the SEM photographs in Figure I.2. The IN concentrations go through an order of magnitude variation. Table 5.2 indicates the variability cannot be explained significantly by any of the aerosol elemental tracers. A slight positive relationship is apparent between IN and Al. The highest IN concentrations occur just after the occurrance of high winds from the interior of Iceland with observations of blowing dust. It is suspected that local blowing dust is an important factor in determining the IN variability. In addition, sample 9 was collected on a local holiday with above normal traffic on local dirt roads. Al and the IN concentrations appear to respond to this local source. One of the lowest IN concentrations, as low as the concentration associated with the background airmass to the north in sample 4, occurred during sample 7, when the pollution aerosol tracers were at a maximum, and haze was observed.

I.2 Winter Study

The winter aerosol study was conducted during the period 16-23 February, 1980. Seven, 24-hour aerosol sample sets were collected and analyzed. The bulk-aerosol trace-element composition is presented in Table 10 in Appendix A. The summary plot of the aerosol elemental tracers is given in figure I.3. Figures I.4a-d are the isohypse analyses for the 850 mb and 700 mb pressure levels for this sampling period. Figures I.5a-d are the SEM photomicrographs of the aerosol particles collected during samples 3 and 5.

During winter, Iceland is located along the storm track in the North Atlantic Ocean. The weather throughout the sampling period was characterized by low clouds, fog, rain and snow with winds typically moderate to strong. Wind speeds aloft were also high. The isohypse analyses in Figure I.4 indicate this by the length of the isohypse between 24 hour tick marks. Wind direction was typically from the southwest, placing the northeastern United States upwind of the site. The isohypse analyses suggest that transport times from the North American source region may have been on the order of 3-4 days. The straight line distance from Irafoss to New York is 4200 km. Even though Iceland is more than twice the distance from the USA as that of Europe, the transport time from the USA pollutant aerosol particle emissions is 1-4 days less than it was from Europe during the summer experiment. This is evident from comparisons of Figure I.4 and Figure 4, Appendix C. Because of the stronger steering winds aloft from the west, surface low pressure centers moved rapidly through the region. This is in contrast to the summer situation. During summer, low centers could stall to the east of Iceland long enough to cause a retrograde advection from east to

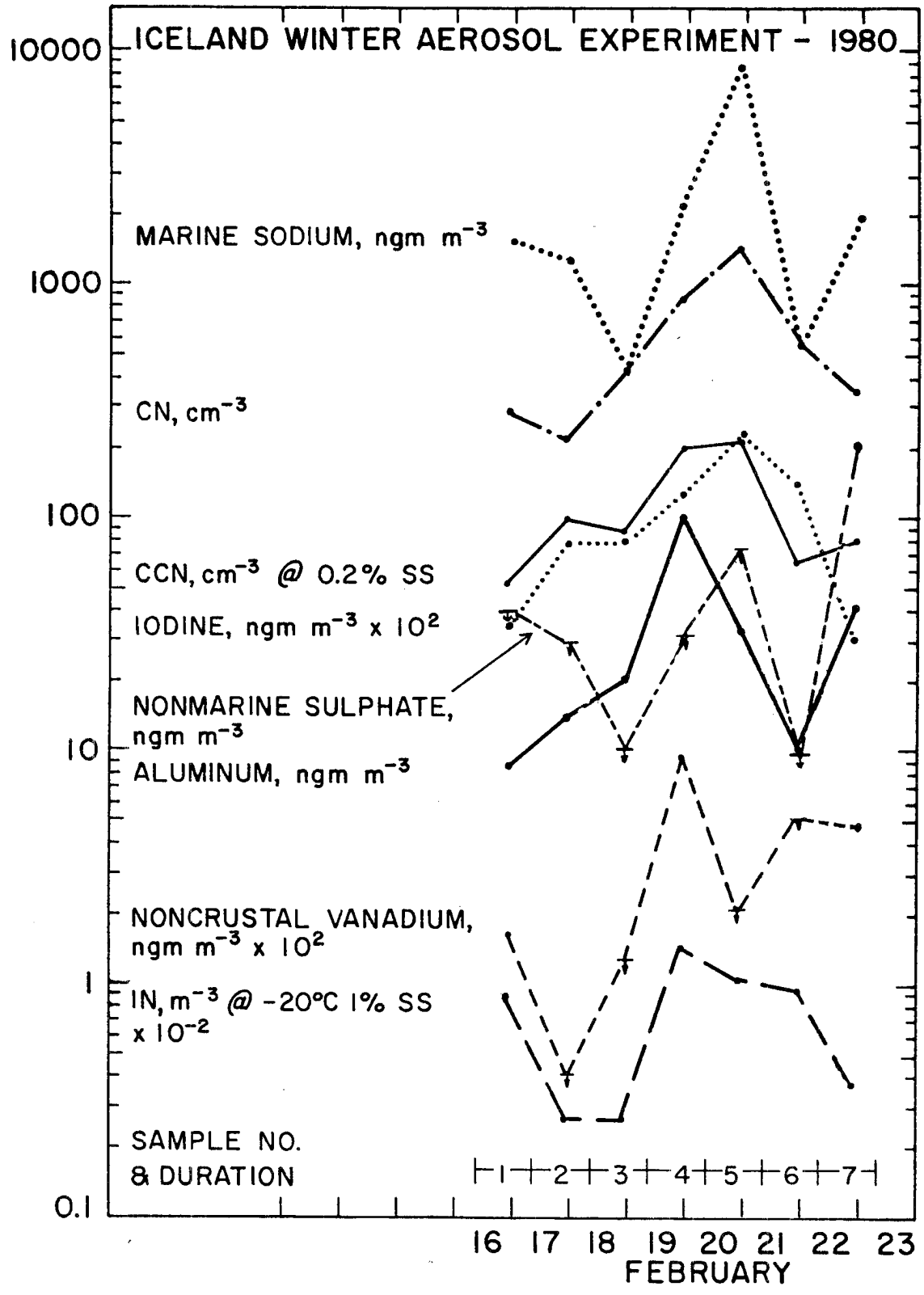


Fig. I.3 Irafoss, Iceland winter aerosol experiment, February 1980.

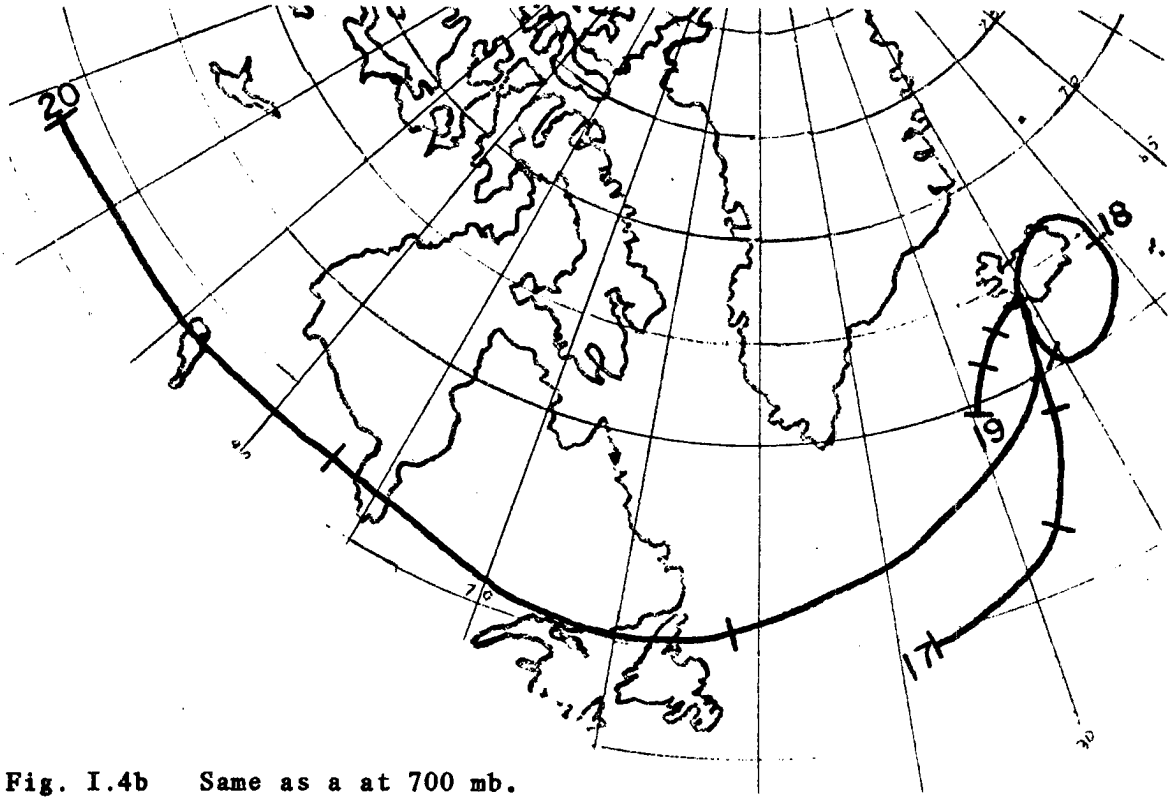


Fig. I.4b Same as a at 700 mb.

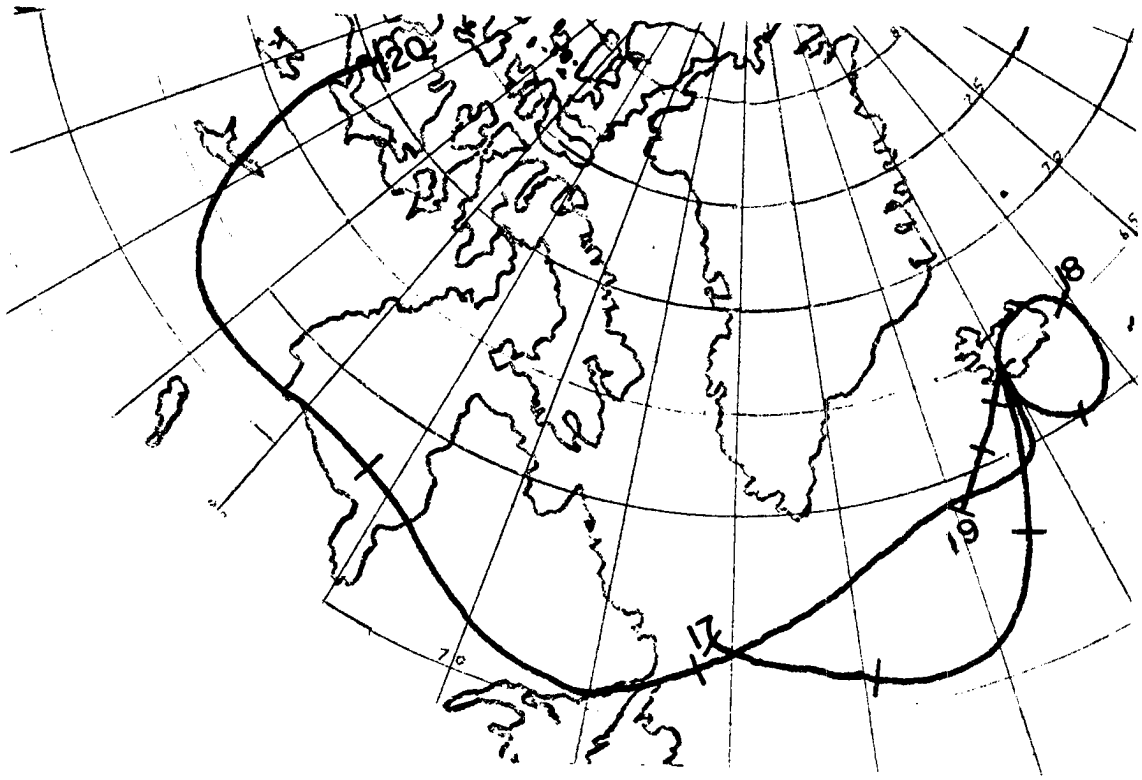


Fig. I.4a Irafoss 850 mb isohypse analyses, 17 - 20 February, 1980, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

Fig. I.4c Same as a at 850 mb, 21 - 23 February, 1980.

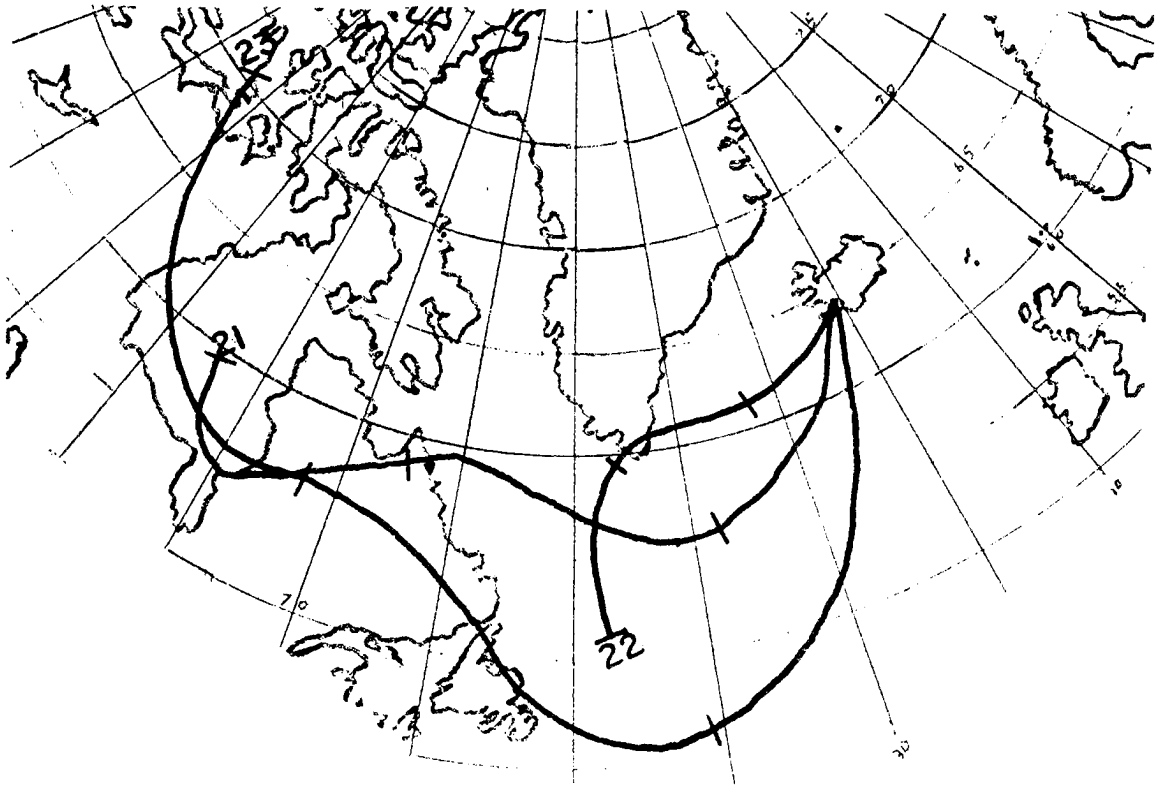


Fig. I.4d Same as a at 700 mb, 21 - 23 February, 1980.

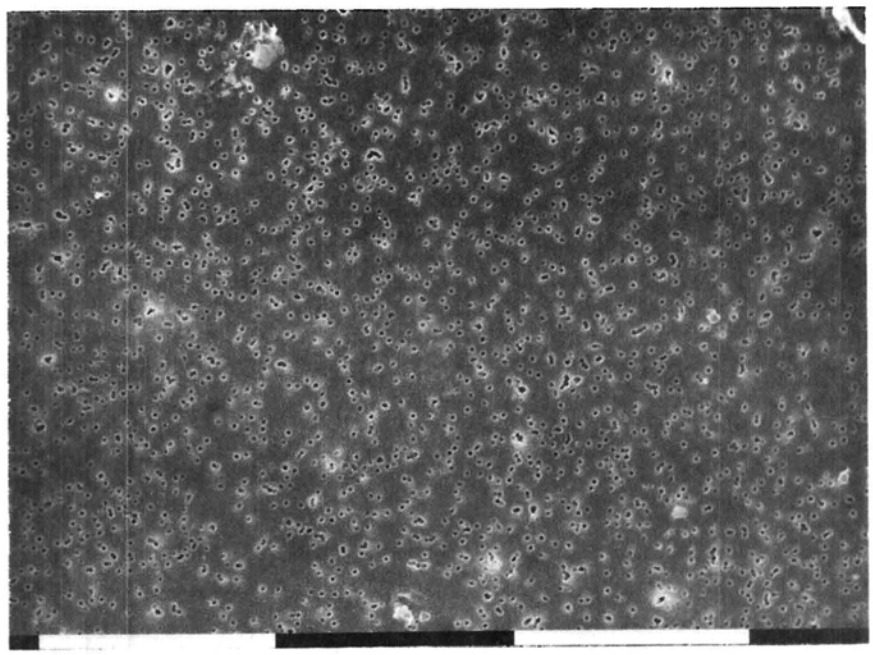


Fig. I.5a Scanning electron microscope photographs of aerosol particles collected at Iraföss during winter on 0.2 μm diameter pore size Nuclepore filters from sample #3, 3100X magnification, bar scale is 10 μm.

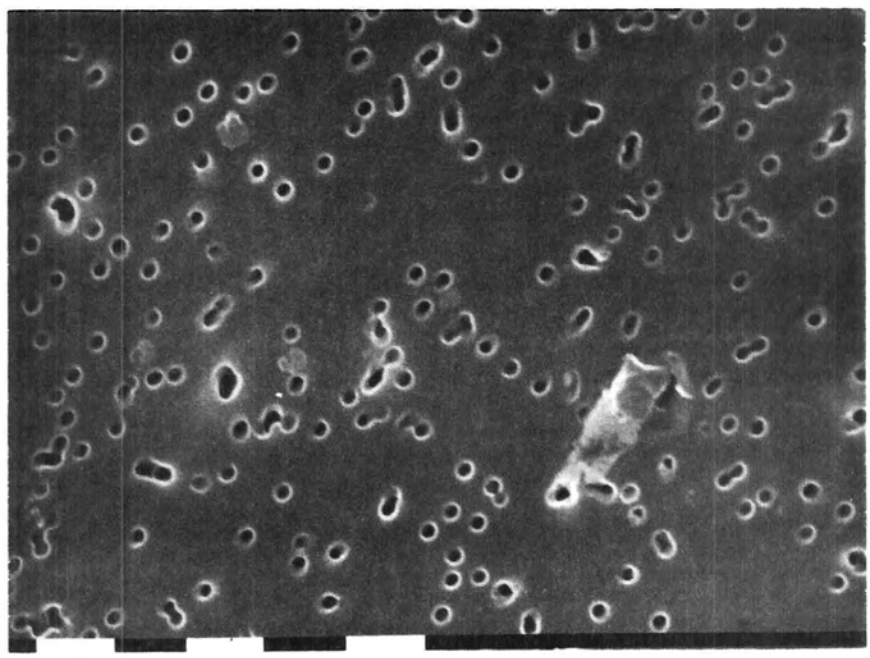


Fig. I.5b Same as a but 10,000X magnification, bar scale is 1 μm.

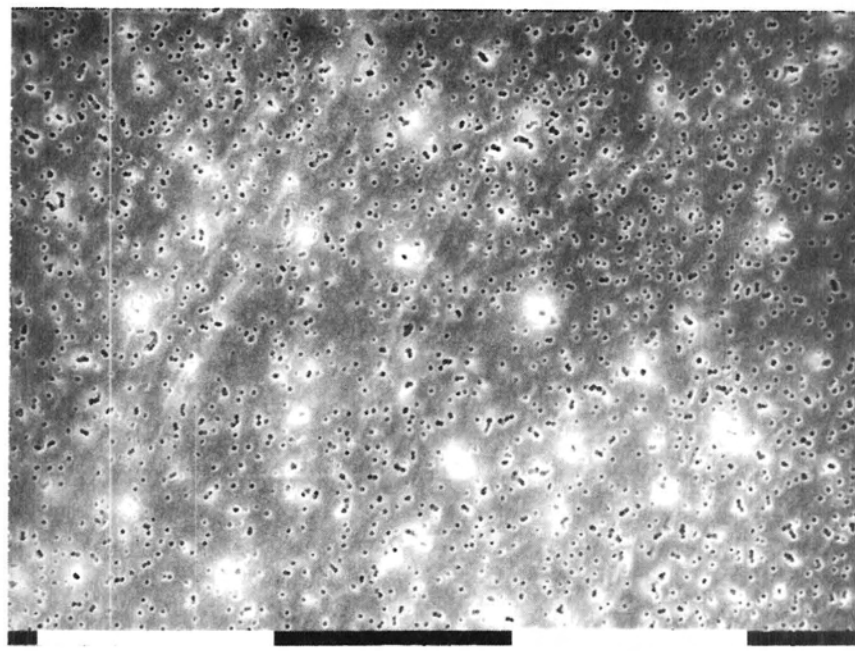


Fig. I.5c Same as a, but for sample #5.

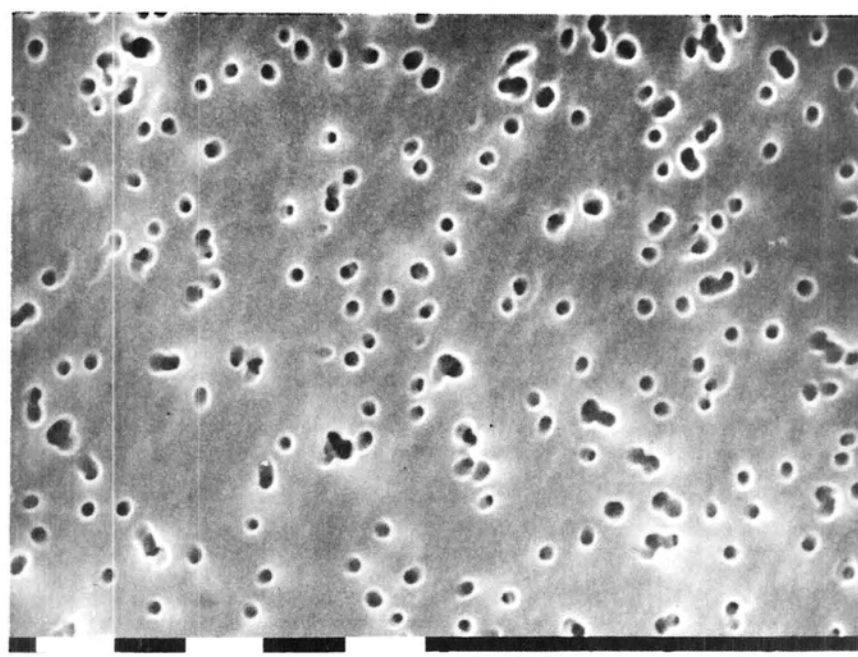


Fig. I.5d Same as c, but 10,000X magnification, bar scale is 1 μm.

west around the low from Europe to Iceland. The rapid movement of lows during winter prevents this from occurring. Flow from the west dominates.

Iceland, during winter, is situated in a stormy maritime environment. As evidenced from the Pangnirtung, NWT, Canada experiment, this stormy region removes aerosol particles efficiently. Since Iceland is in the direct path of the storms, the aerosol concentrations and composition should reflect this. Comparison of Figures I.1 and I.3 shows that this is indeed the case. Concentrations of all the aerosol tracer components, except the marine tracers, are down from their summer values. In the case of the marine tracers, marine Na is 5–6 times higher and I is about the same. The higher sea-salt concentrations reflect the higher mean wind speeds. During the course of the experiment, there was one short wave passage. Its approach and retreat can be seen in the peak of marine Na which occurred during the course of samples 3, 4, 5 and 6. The Al concentration averaged in concentration 15 times less than the concentrations during the summer. However, Al still ranges in concentration over a factor of ten, and peaks on sample 4. Winds during this sample at the surface were south to southeast at 30–35 kts. Even though the ground was wet, these winds apparently were able to loft local soil dust which accounted for the Al peak.

The pollution aerosol tracers V and $SO_4^=$ were very low in concentration and near their detection limits. V was only measureable on four samples. Non-marine $SO_4^=$ was only detected once, in sample 7. These concentrations are as low as those measured at Barrow during the summer. Although Iceland is the closest arctic site to Europe, in terms of distance of transport, and to the USA, in terms of time of transport,

the aerosol at Iceland has the elemental composition of the most remote arctic site, Barrow, during its cleanest time of the year, summer. Iceland is characterized best as a remote maritime site during the winter. The SEM photomicrographs in Figure I.5 show the particles collected in samples 3 and 5. Few particles are present.

I.2a Cloud condensation nuclei

The CCN concentrations at 0.2% SS averaged 105 cm^{-3}. This is about the same as the CCN concentrations during the summer experiment, and typical for maritime conditions. The data in Table 5.2 also confirm that much of the variance of the CCN can be explained by the marine aerosol elemental tracers, marine Na and I. This is similar to the summer case, however, because of the lack of pollutant aerosol during the winter, the variance can be explained by the marine tracers alone. The high covariance of CN with CCN is a result of the clean, background nature of the aerosol at Irafoss. The CN correlate positively with the marine tracers marine Na and I, with correlation coefficients of 0.88 and 0.92 respectively. Thus, the majority of the aerosol numbers are of marine origin as well as their mass. The CN and the CCN concentrations converge to the same values, in the mean, for the entire period. The relatively high covariance of Al and CCN is most likely a result of the simultaneous production of sea-salt particles and local dust as a result of their generation dependence on wind speed.

I.2b Ice nuceli

The IN concentrations at $-20^{o}C$ and 1% WSS were slightly higher during the winter experiment than they were during the summer experiment. Referring to Table 5.2, the IN showed no significant correlation with any of the elemental source tracers. In general, there

was some positive correlation with all of the tracers. The IN appear to track the general rise and fall of the aerosol tracers, which, as previously discussed, are related to the wind speed. Note that the IN have a maximum during sample 4, the same as Al. The corresponding peak of V is not significant because of the low concentration and the resulting uncertainty in its determination. Based upon this sample, the IN are probably most closely linked to local dust. This was also deduced from the summer experiment.

I.3 Seasonal Differences in Cloud-Active Aerosol

Figure I.6 presents the seasonal mean CCN spectra for summer and winter at Irafoss, Iceland. Figures I.7 and I.8 depict the individual IN concentrations for summer and winter with the mean spectra also plotted. There was difficulty in the IN analysis at $-28^{o}C$ due to the high blank filter values. All data for $-28^{o}C$ for the winter experiment were less than blank. The greatest different between the two IN spectra is in their slope. This is typical of the difference between IN spectra from remote areas and areas affected by human activity. Spectra in remote areas have smaller slopes than those from urban areas. The summer experiment at Irafoss was characterized by an anthropogenic aerosol component whereas the summer case was essentially a background aerosol condition.

The concentration of IN at the temperatures of $-16^{o}C$ and $-20^{o}C$ is lower during the summer than during winter. This is similar to the situation in Barrow where the concentrations of IN are inversely related to the anthropogenic character of the aerosol. The effect in Iceland may not be as pronounced due to the lower contrast between the winter and summer aerosol population. The higher winter concentrations of IN

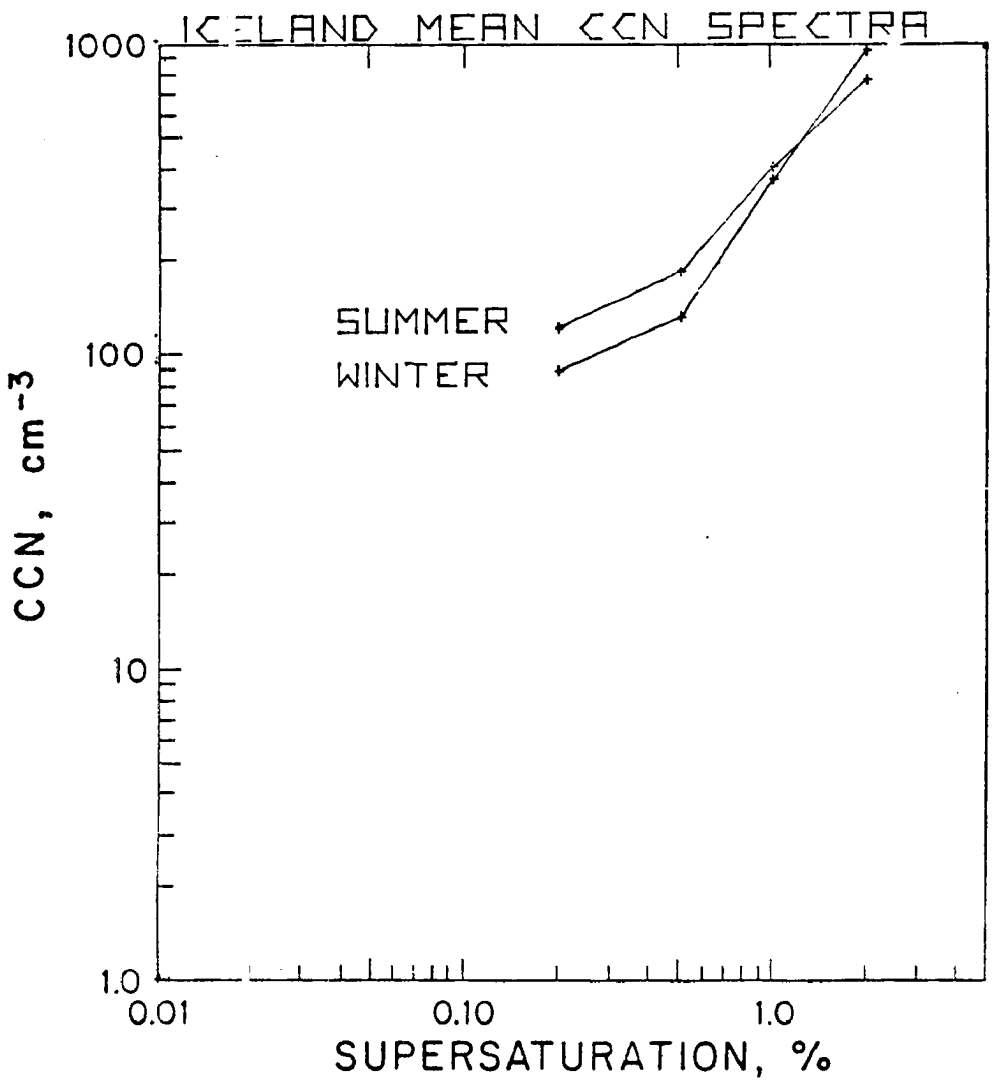


Fig. I.6 Mean CCN spectra for Irafoss, Iceland, derived from nineteen spectra measured during summer, 14 - 25 August, 1979 and fifteen spectra measured during winter, 16 - 23 February, 1980.

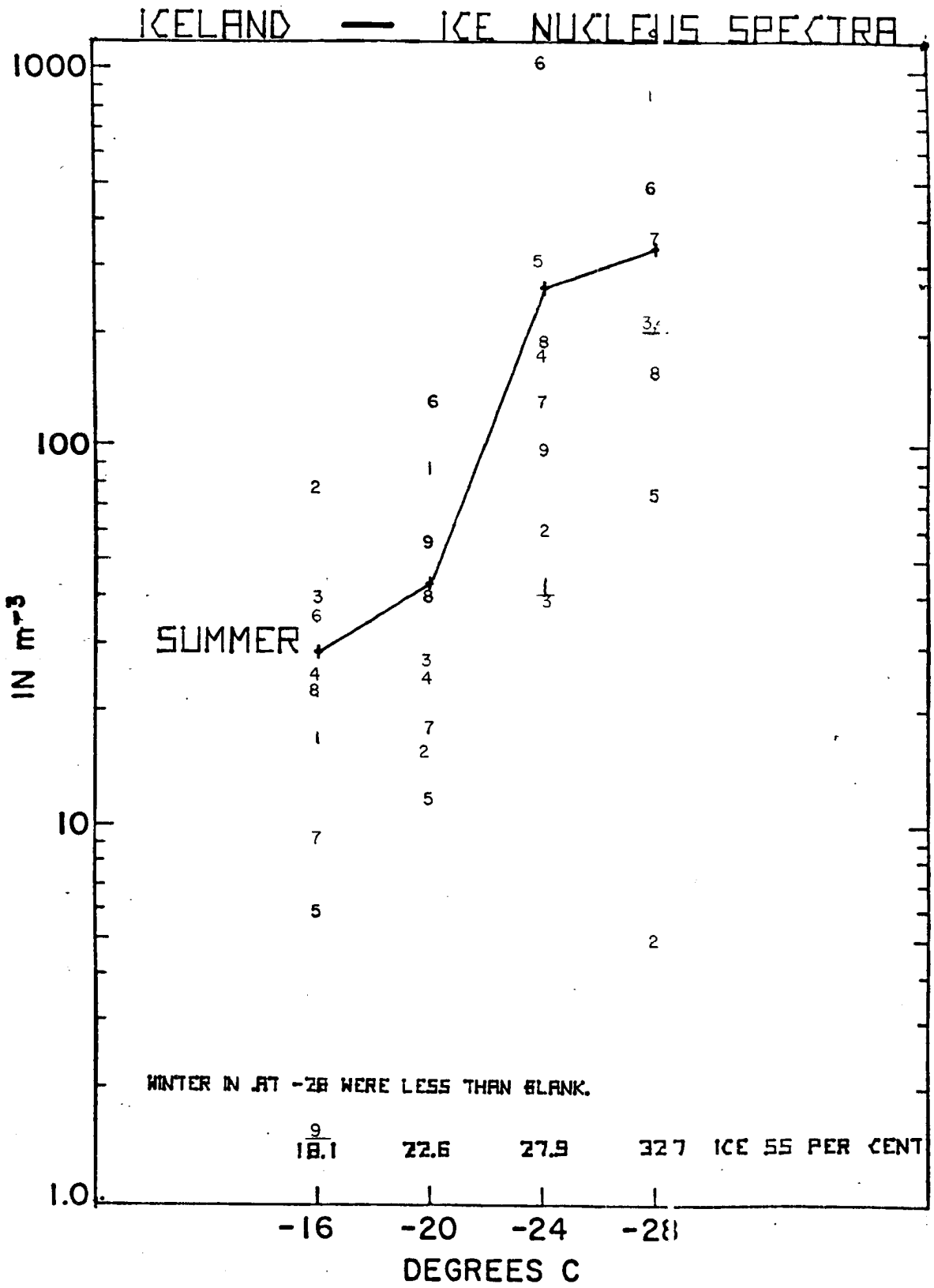


Fig. I.7 IN spectra for Irafoss, Iceland for the summer experiment 14-25 August 1979. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

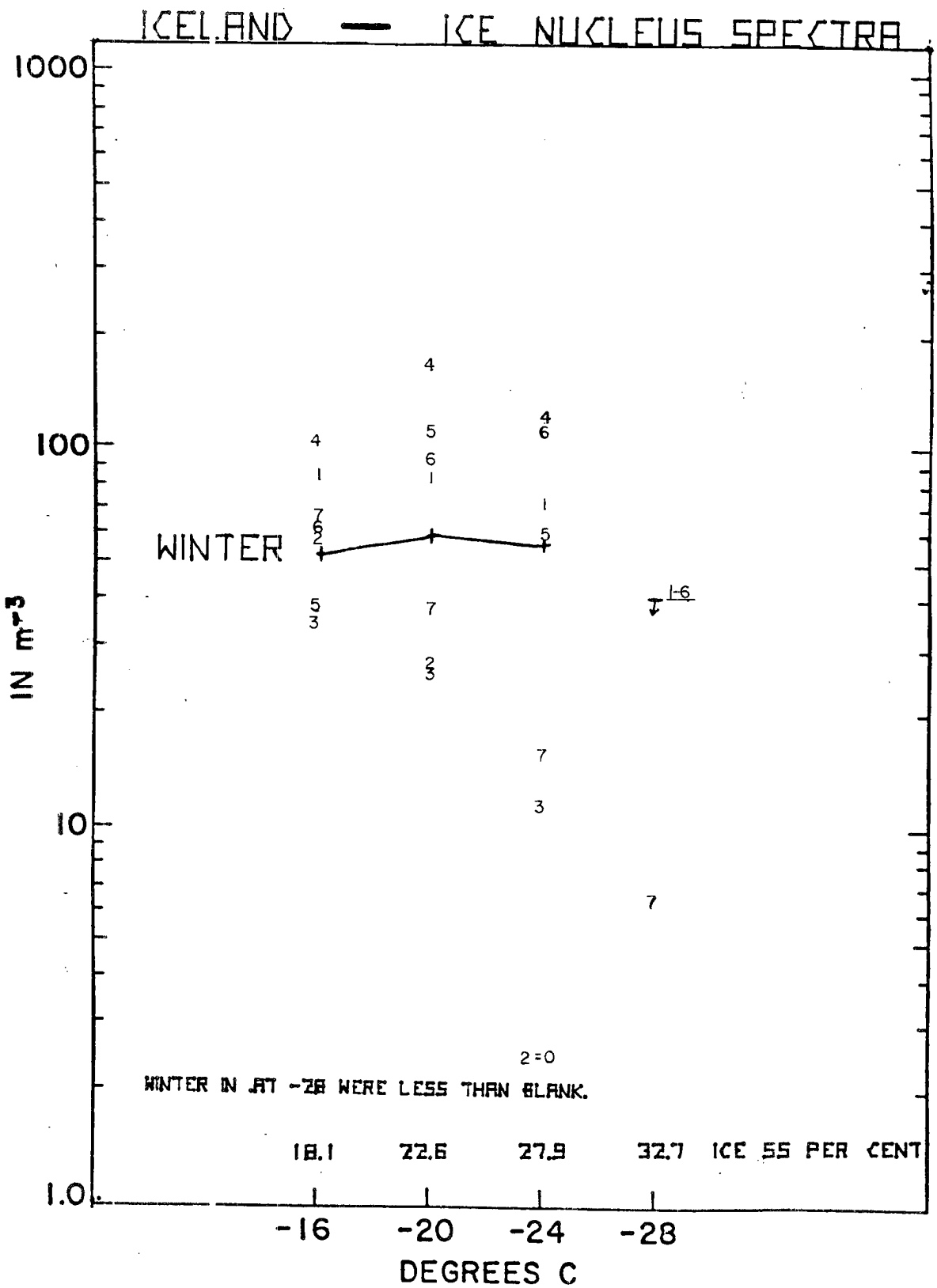


Fig. I.8 IN spectra for Irafoss, Iceland for the winter experiment 16-23 February 1979. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

occur even in light of the fact that the suspected local dust source as indicated by Al is 15 times higher during summer. The advection of anthropogenic aerosol particles from Europe during summer caused an increase in aerosol loading, including Al, but decrease in the IN concentration. Thus the reverse seasonal behavior of the IN at Irafoss, as compared to the IN behavior at Barrow, is apparently due to the reverse behavior of the anthropogenic character of the aerosol between these two sites. The aged pollutant aerosol present at Irafoss during part of the summer experiment had greater mass and number concentration than the background aerosol of summer and winter but fewer IN amongst the total aerosol. This suggests a "poisoning" effect of the pollutant aerosol particles and gases on the IN in the aged (> 5 days) polluted airmass that was studied at Irafoss during the summer. This does not support the idea that the IN are selectively removed by activation in the atmosphere as a result of colder temperatures. Summer temperatures averaged 6°C warmer than the temperatures during the winter experiments (Table 5.1), yet the IN concentrations were highest during winter (Table 5.4).

The same reasoning can be used to explain the slight difference between the winter and summer CCN concentrations at Irafoss. Because of the dominance of the marine aerosol the effect of the pollutant aerosol on the seasonal difference in CCN is small. The effect of the pollutant aerosol is more important at lower supersaturations, as has been the case at the other study locatons where the pollutant aerosol signal is stronger, Barrow, Narragansett and Ny Alesund-YMER. Tables 5.3 and 5.4 also suggest this interpretation by comparing the mean winter/summer ratios of the IN at -20°C and 1% WSS and the CCN at 0.2% SS with the

pollutant indicators noncrustal V and nonmarine $SO_4^=$. These seasonal differences are slight but when interpreted with the meteorology, transport and removal processes, and the local site conditions, the above interpretation emerges.

I.4 Sources of Cloud Active Aerosol Sources

The major sources of cloud-active aerosol at Irafoss, Iceland are a combination of both natural and anthropogenic, depending upon the season. The IN showed no dependence upon anthropogenic emissions as a possible source. Conversely, the summer experiment showed an inverse relationship of IN concentration to the presence of anthropogenic aerosol. For both seasons, there appeared to be a dependence on local dust evolution. This could be related to both the natural wind and the activities of man (road dust). The most important conclusion was the inability to detect a positive dependence of the IN at this site upon the long range transport of aerosol particles from any pollution source region.

The CCN concentrations on the other hand show a strong dependence upon long-range pollution transport. In this case, the source region was industrialized Europe to the east. No effect could be detected resulting from emission and transport of pollutants to Irafoss from North America. When pollution aerosol was not present at the site, during the winter, the CCN could be explained by the presence of natural marine primary and secondary aerosol. Therefore, under the proper conditions, pollutants advected to this site play an important role in determining the CCN concentrations. The conditions necessary for this to occur are specific. These conditions are unlikely to occur during winter, and only sporadically during the summer. The majority of the

time the CCN at this location are a result of natural processes, with occasional concentration increases resulting from pollutants advected to the site.

APPENDIX J

DETAILED FIELD EXPERIMENT DISCUSSION
Ny Alesund-YMER 80

These two aerosol experiments were conducted in the high arctic. Ny Alesund is a weather station operated by Norway on the island of Spitsbergen at approximately 79° N. The nearest potential source of contamination is the town on Longyearbyen, approximately 100 km to the southeast. Coal mining is done on the island. Coal is used for power production. YMER-80 is the name of an expedition sponsored by the Swedish government to commemorate the 100th anniversary of the voyage of the Vega, a Swedish ship, through the Northwest Passage from Europe to Alaska. The YMER is an icebreaker which was outfitted for aerosol and gas sampling. The YMER expedition sailed in the vicinity of the islands of Spitsbergen and Greenland during the summer of 1980. Great care was taken to avoid contamination from the ship, which was the major potential source of contamination. This area was chosen as a study location because of its position along potential transport pathways from both Europe and North America. This would allow the comparison of the relative importance of these two source regions on the total aerosol advected to the north via this meteorological gateway to the Arctic.

J.1 Summer Study - YMER-80

The summer aerosol experiment was conducted aboard the YMER during the period 11 August to 1 September, 1980. Eleven sample sets were collected and analyzed. These samples extended over 1-3 day periods.

The samples were taken only when there was sufficient evidence of no ship contamination and were generally restricted to periods when the ship was underway. The samplers were located on a forward deck of the ship, ahead of any exhaust ports. Extensive studies had been performed to assure the location was adequate for the purpose of aerosol particle sampling. The actual cruise track and the segments of the track along which the samples were collected is shown in Figure J.1. The numbers between tick marks along the cruise track denote the number of the sample collected along that portion of the cruise track. Figure J.2 is a plot of the sample elemental source tracers and cloud active aerosol by sample number corresponding to the map location and date. The total bulk-aerosol trace-element composition is given in Table 11, Appendix A. Figures J.3a-d are SEM photomicrographs of aerosol particles collected during samples 3, 5 and 10. Figures J.4a-w are surface, 850 mb and 700 mb two day backward isobaric trajectory analyses plotted for the ship's position at 0Z for each day of the aerosol experiment. These trajectories were supplied by Dr. Lothar Schutz of the Institute for Meteorology, University of Mainz, Mainz, Fed. Rep. of Germany.

The aerosol experiment can be divided into two sections. The first section consists of samples 1-3 when the ship was south of the polar front and within an airmass originating from the south. The second section consists of the remaining samples 4-11 when the ship was north of the polar front and within the summer arctic airmass. Note that sample three of the first section was collected at 82^{o} N, under the influence of southern air. The isobaric trajectory analyses in Figure J.4 show the flow from the south during the first three samples.

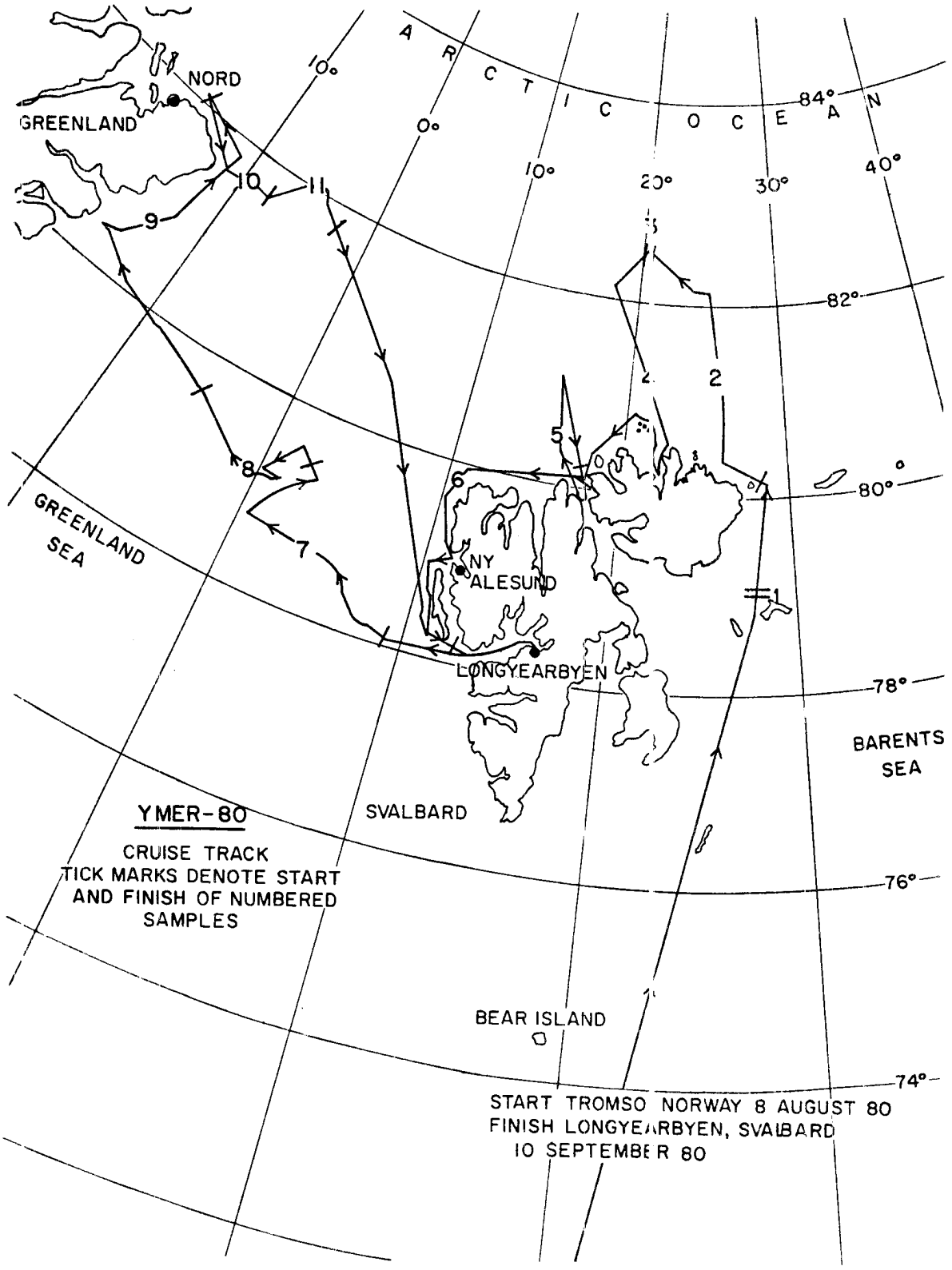


Fig. J.1 YMER-80 Cruise track, Greenland - Svallbard environs, August - September 1980. Numbers along track correspond to sample numbers in figure 5.41. Tick marks surrounding numbers encompass ship's path during the sample.

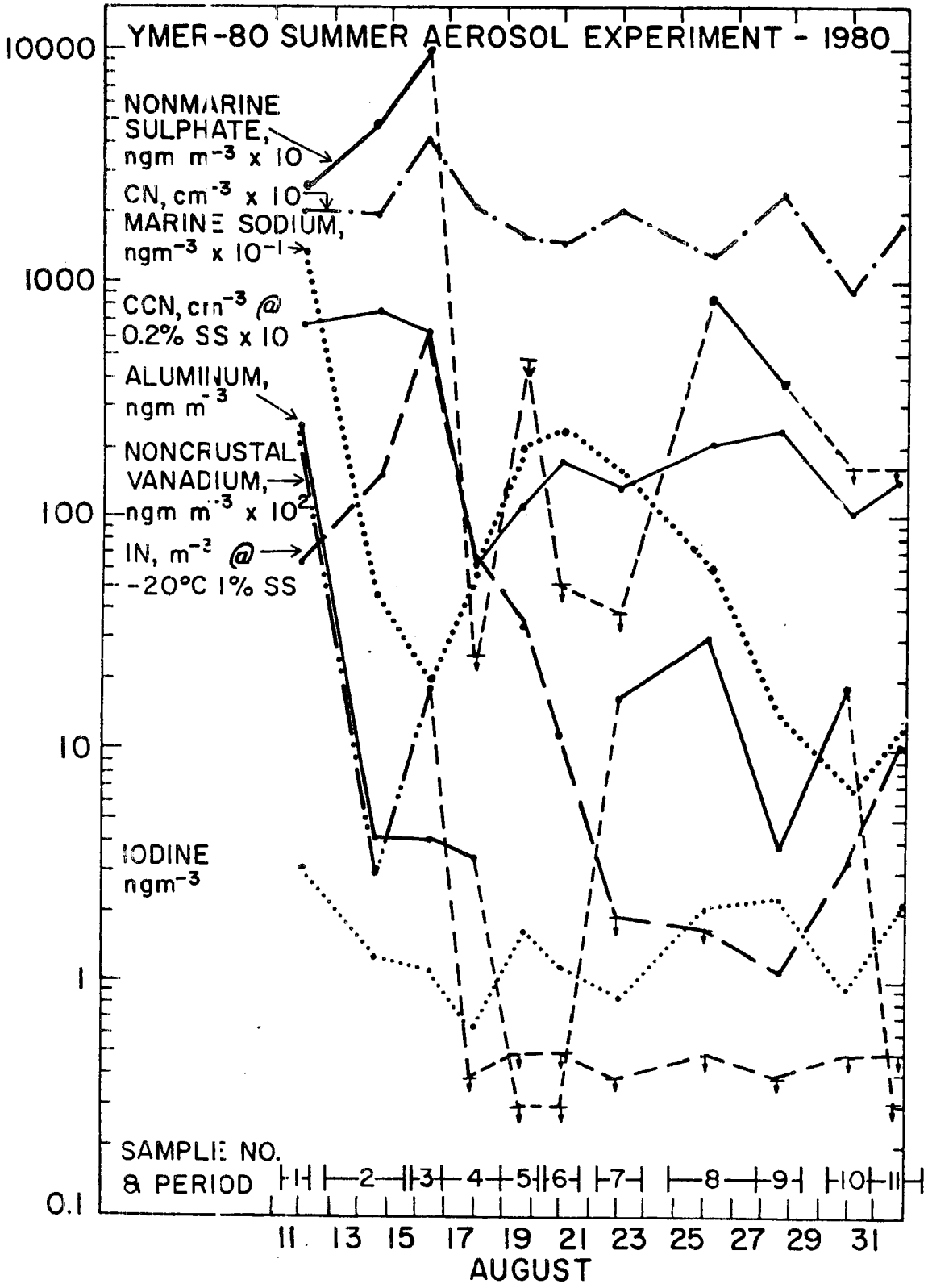


Fig. J.2 YMER-80 shipboard aerosol experiment, August - September 1980.

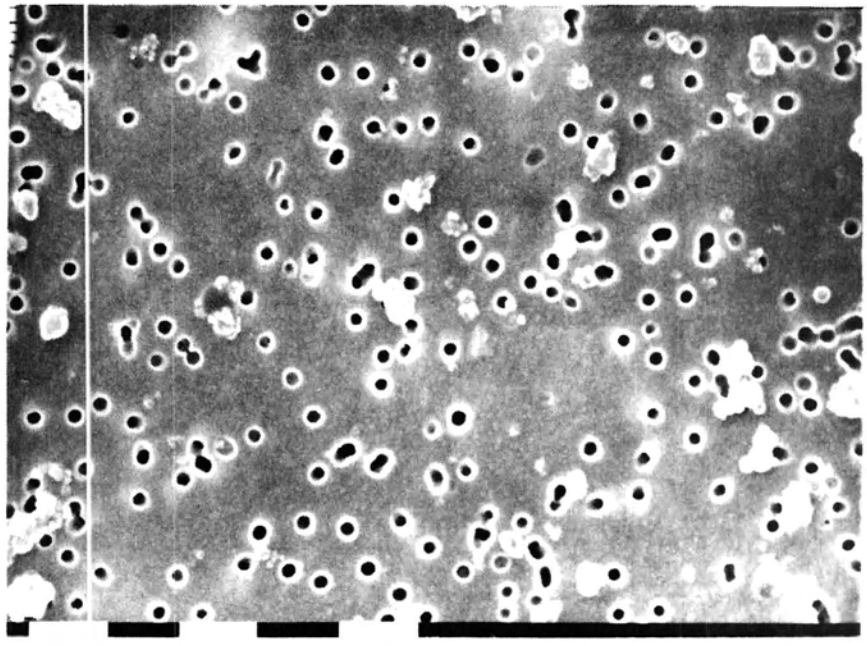


Fig. J.3a Scanning electron microscope photographs of aerosol particles collected at YMER-80 during summer on 0.2 μm diameter pore size Nuclepore filters from sample #3, 3100X magnification, bar scale is 10 μm.

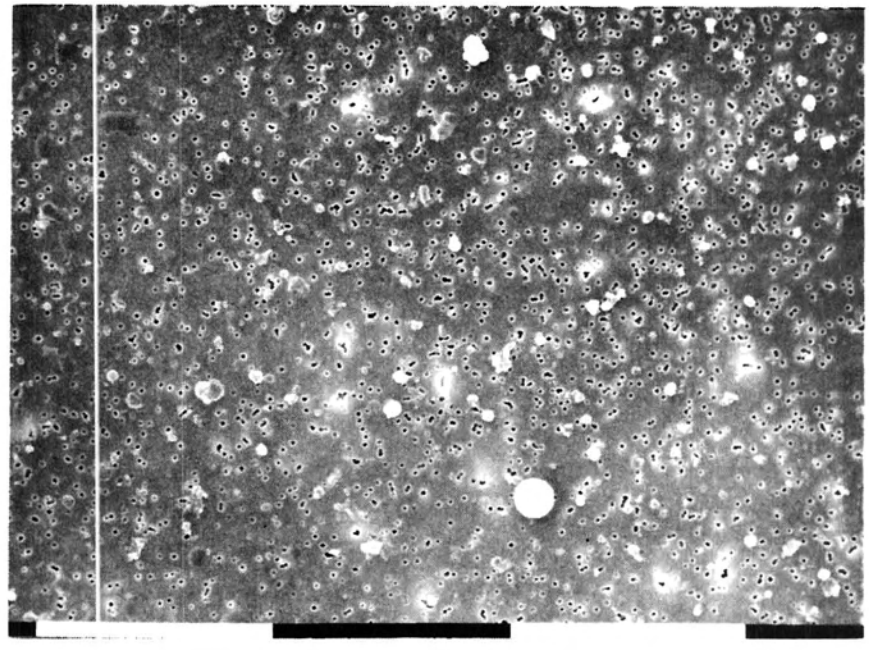


Fig. J.3b Same as a but 10,000X magnification, bar scale is 1 μm.

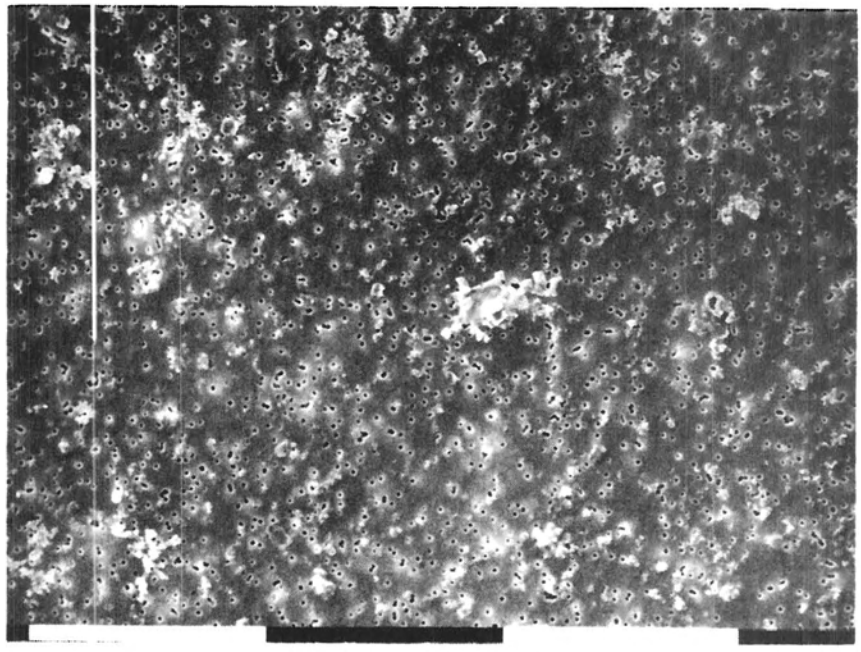


Fig. J.3c Same as a, but for sample #5.

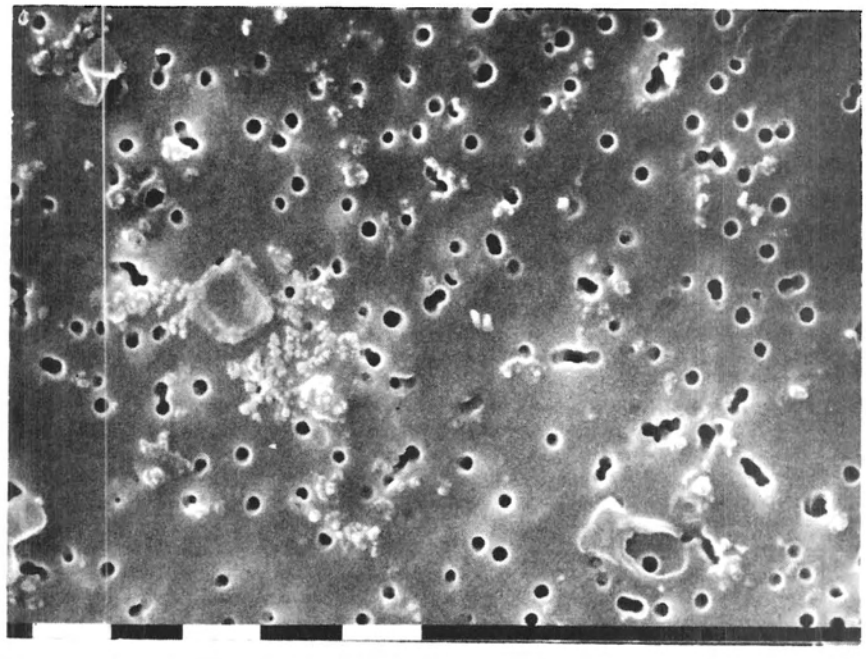


Fig. J.3d Same as c, but 10,000X magnification, bar scale is 1 μm.

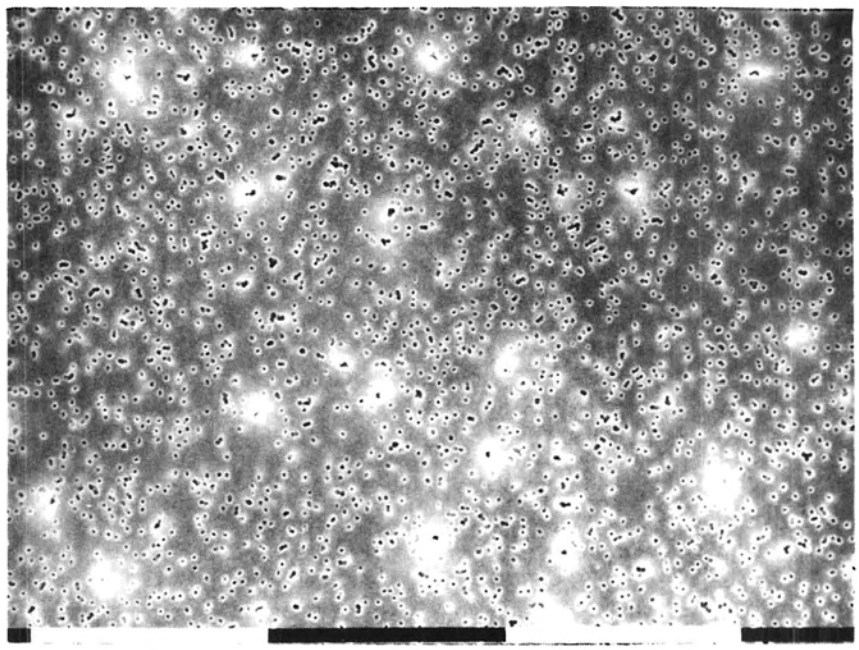


Fig. J.3e Same as a but for sample #10.

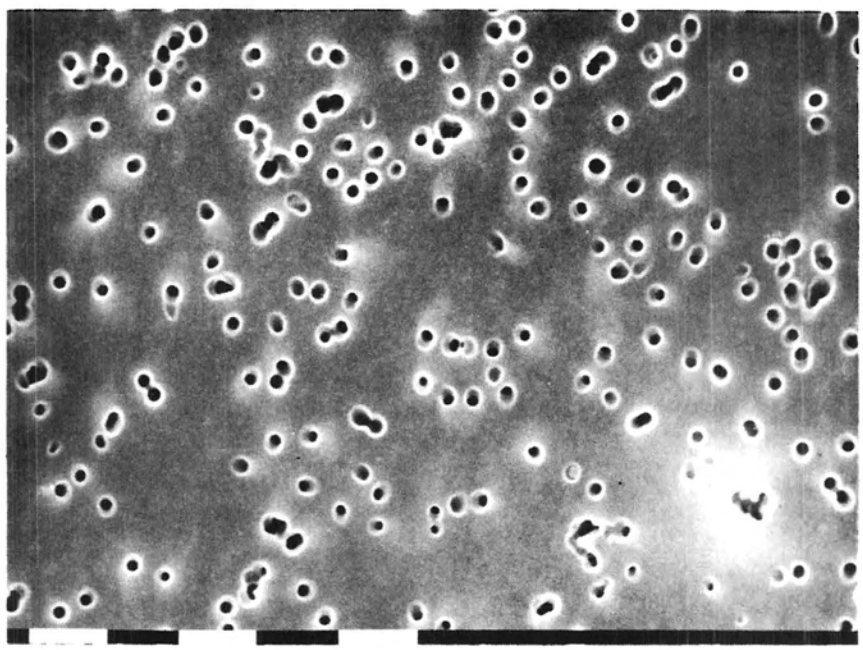


Fig. J.3f Same as e, but 10,000X magnification, bar scale is 1 μm.

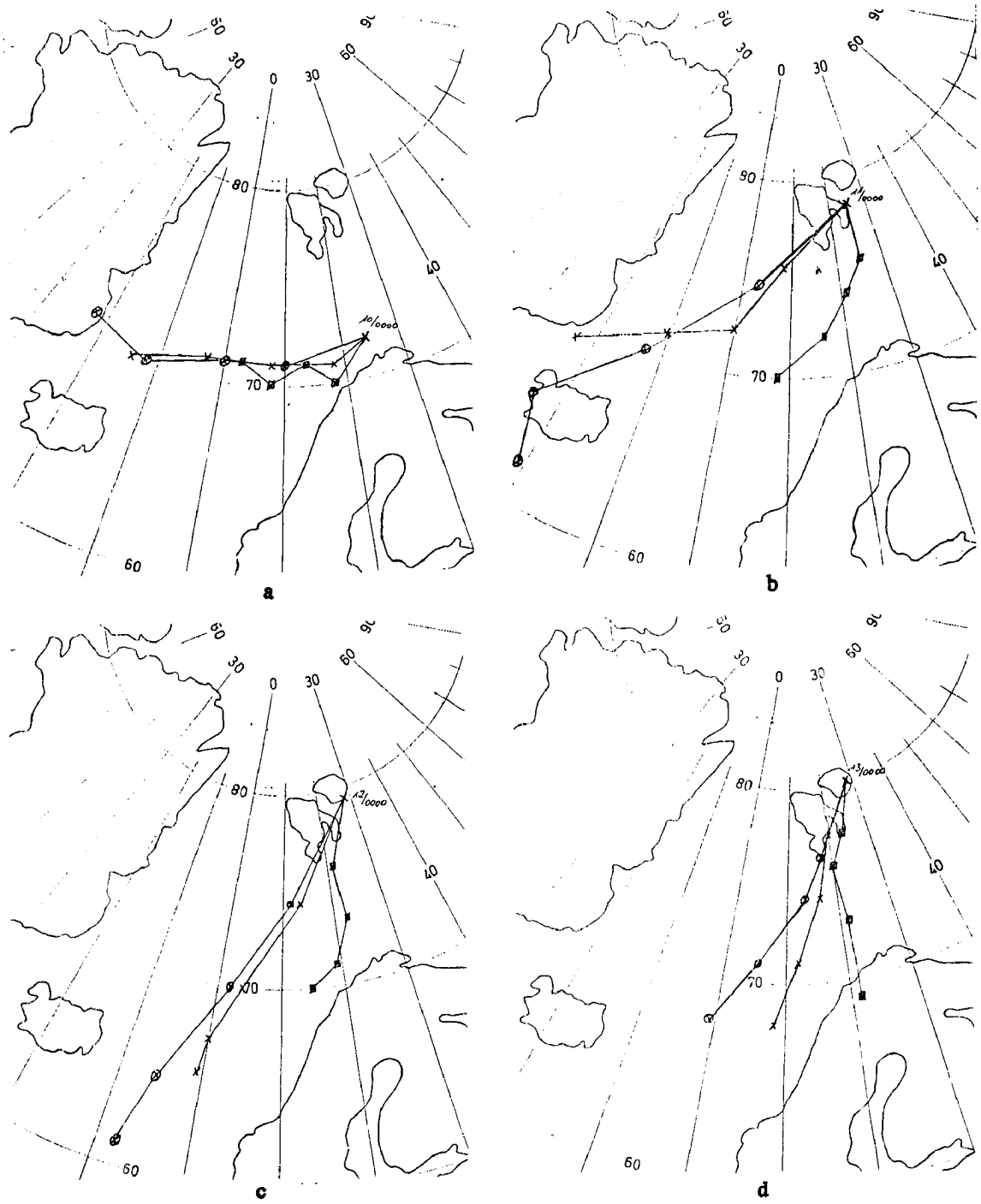


Fig. J.4a-d YMER-80 two-day backward isobaric trajectory analyses for the surface (⊠), 850 mb (X) and 700 mb (⊗) levels, for the ship's position at 00Z in 12 hr. time steps. Date (August, 1980) of trajectory is indicated at ship's position.

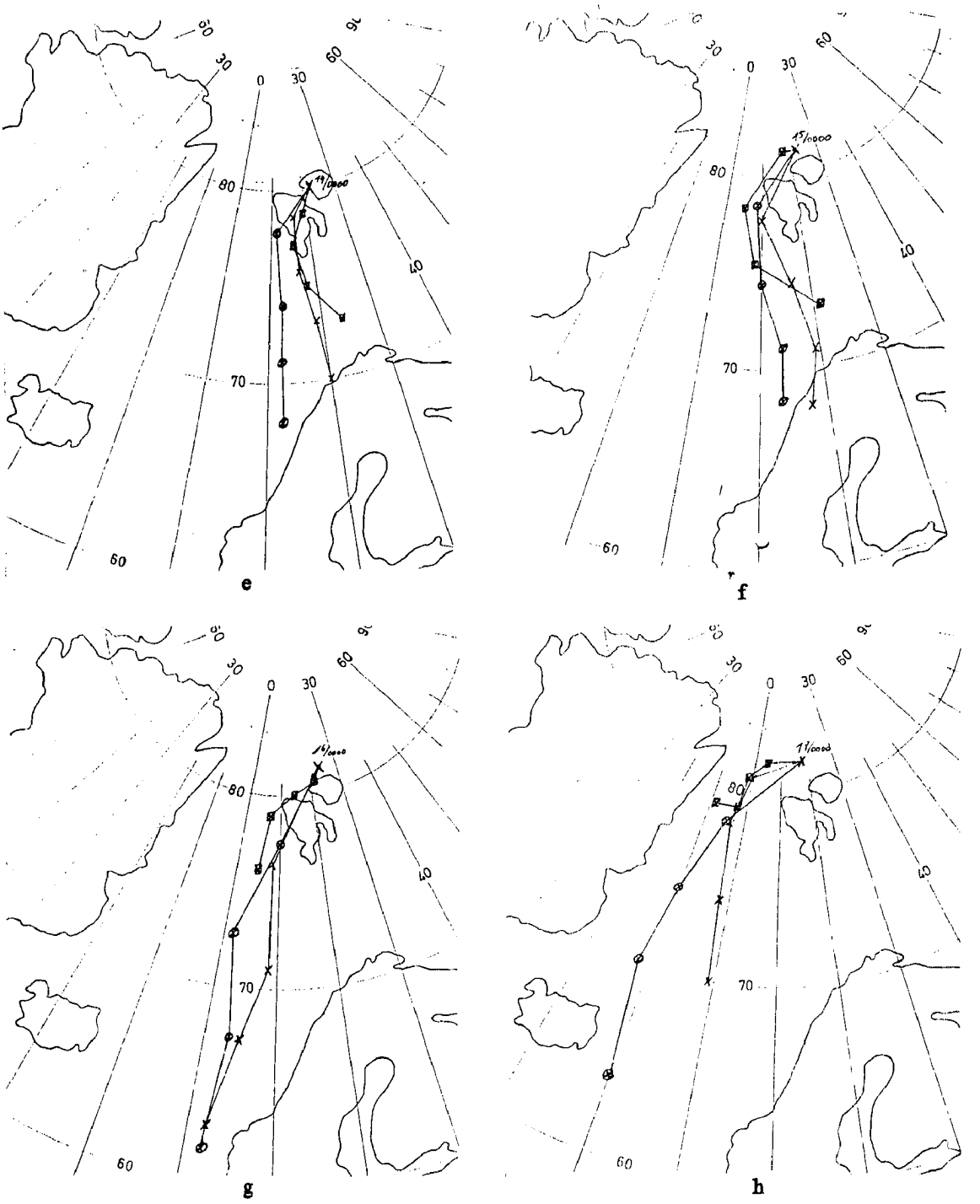


Fig. J.4e-h. Cont.

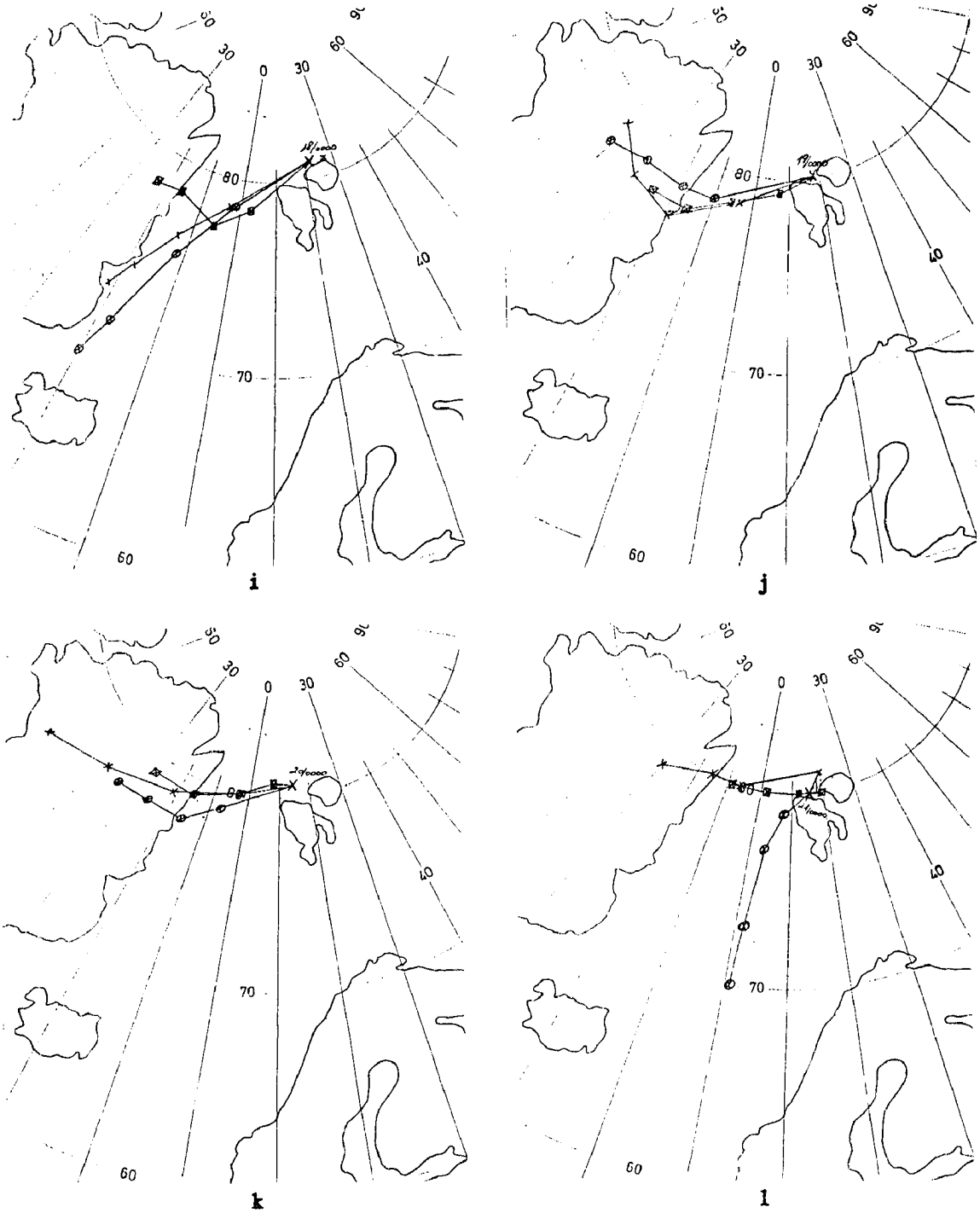


Fig. J.4i-1. Cont.

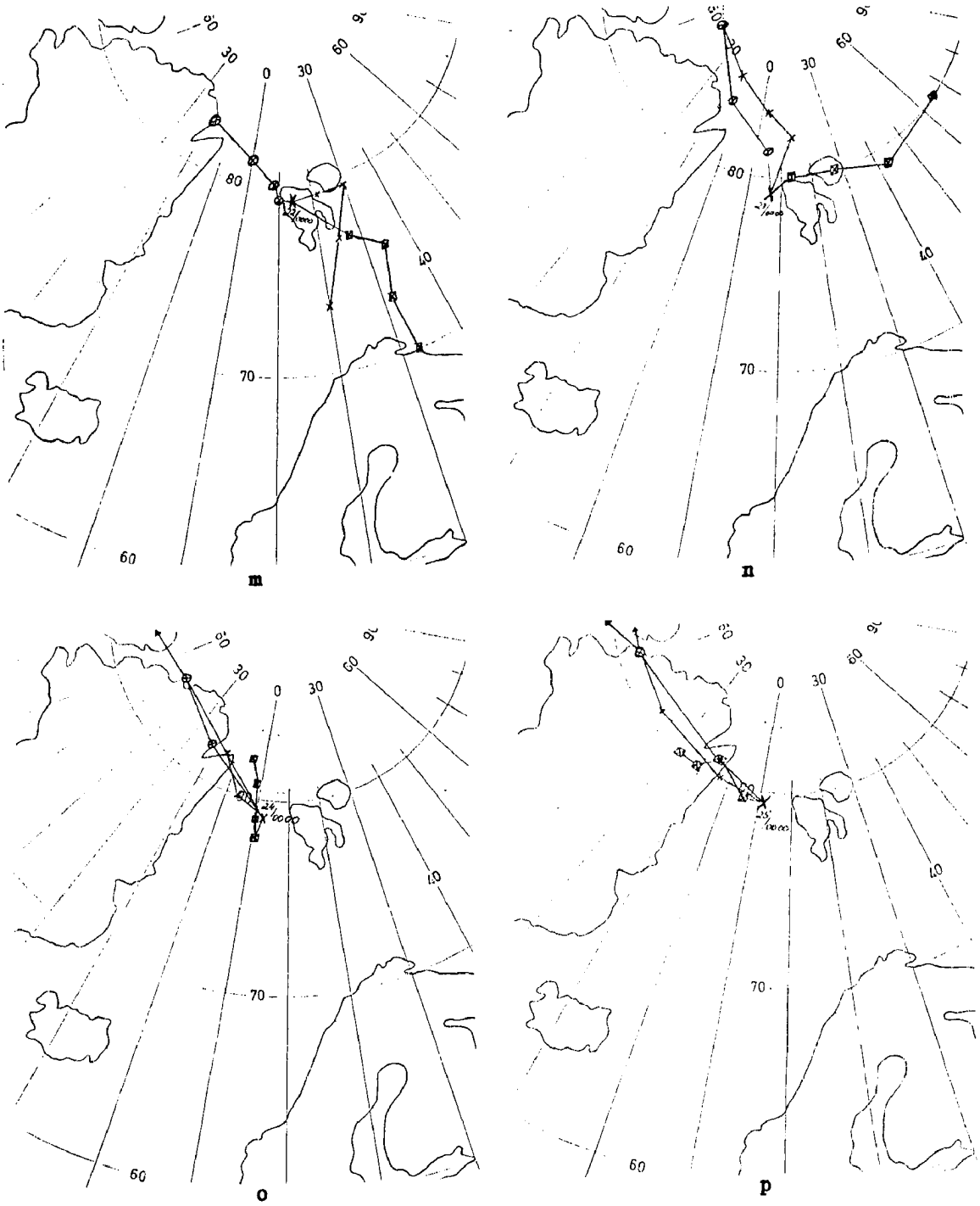


Fig. J.4m-p. Cont.

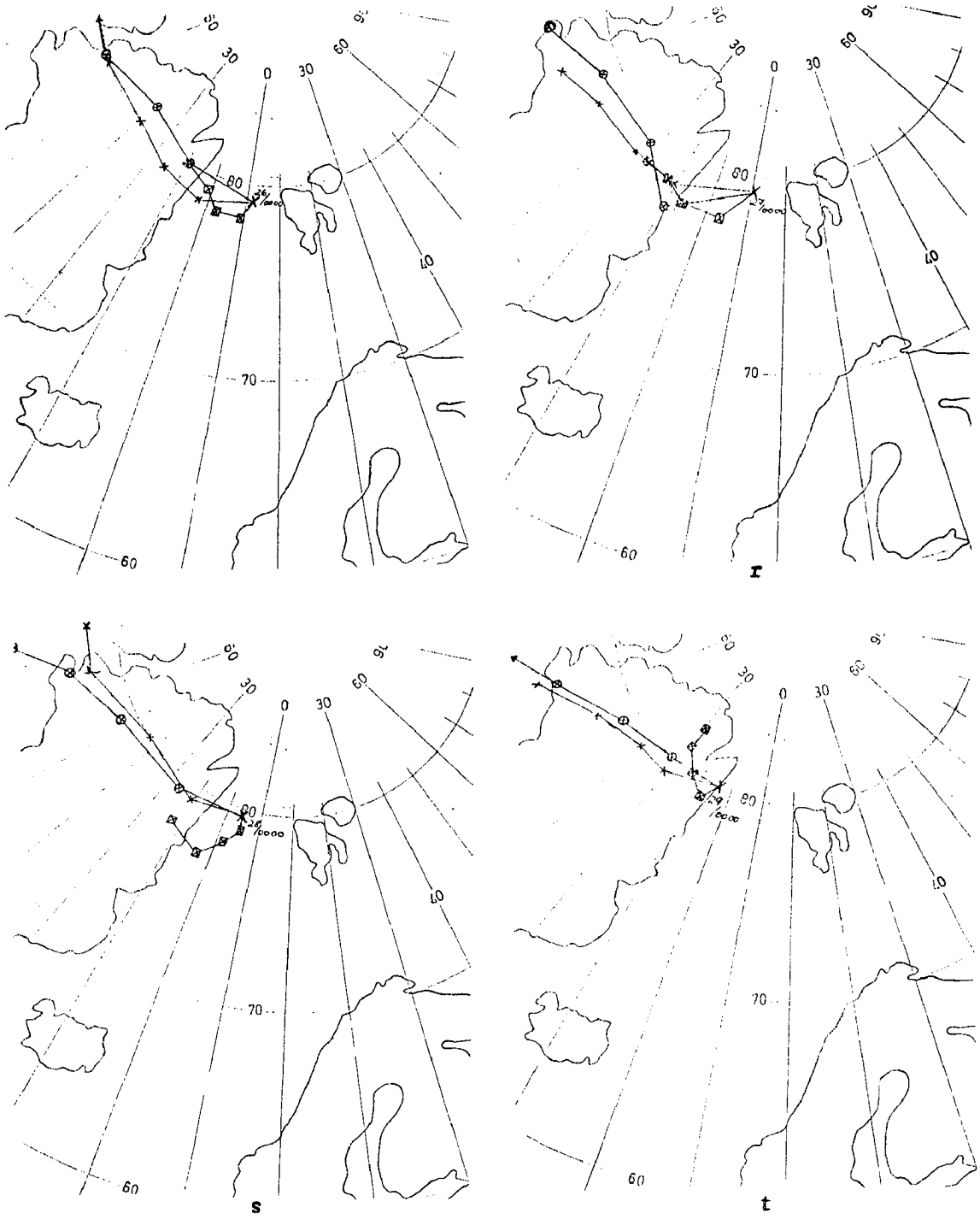


Fig. J.4q-t. Cont.

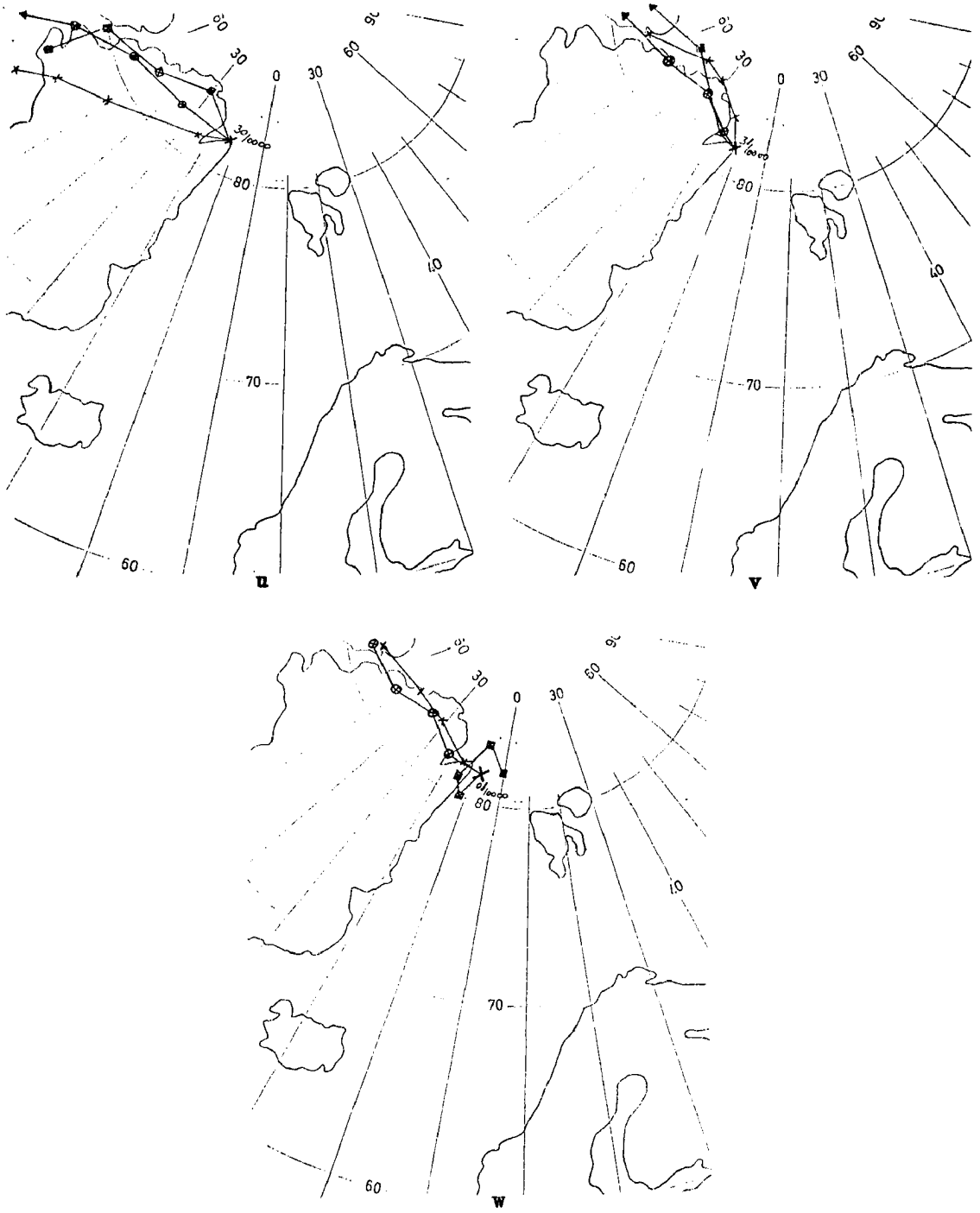


Fig. J.4u-w. Cont. W is valid for 1 September, 1980.

J.1a Cloud condensation nuclei

The CCN at 0.2% SS range over an order of magnitude in concentration during this experiment. Most of this is a result of the first three samples. The average CCN concentration of the first three samples is 67 cm^{-3}, and for the last seven samples, 14 cm^{-3}. The latter concentration is the lowest average measured at any of the arctic sites. The reason for this is probably due to the fact that the ship was primarily operating either within the ice pack or very near its southern limit. After entering the arctic airmass, almost all sources of aerosol particles were cut off. The sea surface was covered by ice, limiting any marine aerosol production. The polar front acted as a boundary to advection of mid-latitude aerosol sources. Low wind speeds also inhibited any mechanical aerosol production mechanisms. Fog was present much of the time. Observations of the CN concentrations as the ship cruised in and out of fog banks suggest the fog acted as an efficient remover of the total aerosol number, presumably by condensation on the fog nuclei and then removal of the unactivated aerosol by Brownian coagulation. This would serve to increase the aerosol particle size and therefore the sedimentation rate. Also it was noted that during subfreezing conditions, rime ice deposits were observed on the pack ice. Thus, there was a near surface mechanism for fog water removal , and therefore aerosol removal, without precipitation. The surface pack ice may act as an aerosol particle sink. These measurements were made further north than any of the other arctic sites, and are the best representation of the aerosol over the pack ice.

Potential for ship contamination always existed. The effect of the ship on the CCN however, was not important. Sample 5 was contaminated

by the ship. This can be seen from Figure J.3c and d, SEM photomicrographs of the aerosol particles from this sample. Note the presence of soot particles on these filters. The CCN were at one of the lowest concentrations at this time. Figure J.2 shows that the pollutant tracers were also not affected by the ship effluent.

Table 5.2 indicates the CCN were significantly correlated with the secondary pollutant aerosol tracer $SO_4^=$. Values of zero were used for samples with less-than values plotted in Figure J.2. The reason for this strong positive correlation is entirely a result of the first three samples taken to the south of the polar front in a mid-latitude airmass. This is not truly representative of the arctic in this region during summer. If samples 1-3 are ignored and a correlation matrix is made for CCN with the natural aerosol tracers Al, marine Na and I and the total aerosol number, CN, the percent variance of the CCN explained by these tracers becomes 4, 0, 53 and 0, respectively. The value for I, 53%, is significant at the 95% level. The true arctic airmass samples indicate the CCN are related to the secondary marine aerosol tracer.

J.1b Ice nuclei

The IN concentrations at -20^oC ranged over almost three orders of magnitude during this experiment as seen in Figure J.2. The highest concentrations occurred when the ship was within the airmass with origins to the south. As a result, the strongest positive correlation occurs between IN and the pollutant aerosol tracer $SO^{4=}$. The highest concentration of both IN and $SO_4^=$ occurred during sample 3 at 82^o N. The non-marine $SO_4^=$ measured at this time can best be attributed to the combustion of coal. Figure J.3a and b show that fly ash particles were collected during this sample. It is apparent that this pollution

aerosol is responsible for the IN concentrations measured. It is possible, based upon the trajectory analyses, that this aerosol originated from the coal burning on Spitsbergen. However, measurements by other investigators on the ship indicated a well aged aerosol and high concentrations of radon daughters were present. The latter is an indicator of the continentality of the airmass as aresult of radon gas sources in rock and soil and gradual radioactive decay of this gas to daughter products which attach themselves to aerosol particles. The airmass appeared to have been advected to the ship from the Eurasian continent to the south. The aerosol collected may be a mixture of these two sources.

Eliminating samples 1-3, the IN show no relationship with any of the measured aerosol source tracers. Concentrations were very low during samples 4-11. The average concentration of IN at $-20^{o}C$ for the last 8 samples was 16 m^{-3}, the lowest concentration measured at any arctic site during this study. This reflects the lack of aerosol sources in this region at this time of year. The ship contaminated filter from sample 5 showed no influence from the ship. The measurements should be representative of the regional aerosol.

J.2 Winter Study - Ny Alesund

The winter aerosol experiment was conducted during the period 29 February to 11 March, 1980. Eleven, 24-hour aerosol sample sets were collected and analyzed. The results of the bulk-aerosol trace-element composition are presented in Table 12, Appendix A. The summary plot of the aerosol elemental tracers is given in Figure J.5. Figures J.6a-d are SEM photomicrographs of aerosol particles collected during samples 4

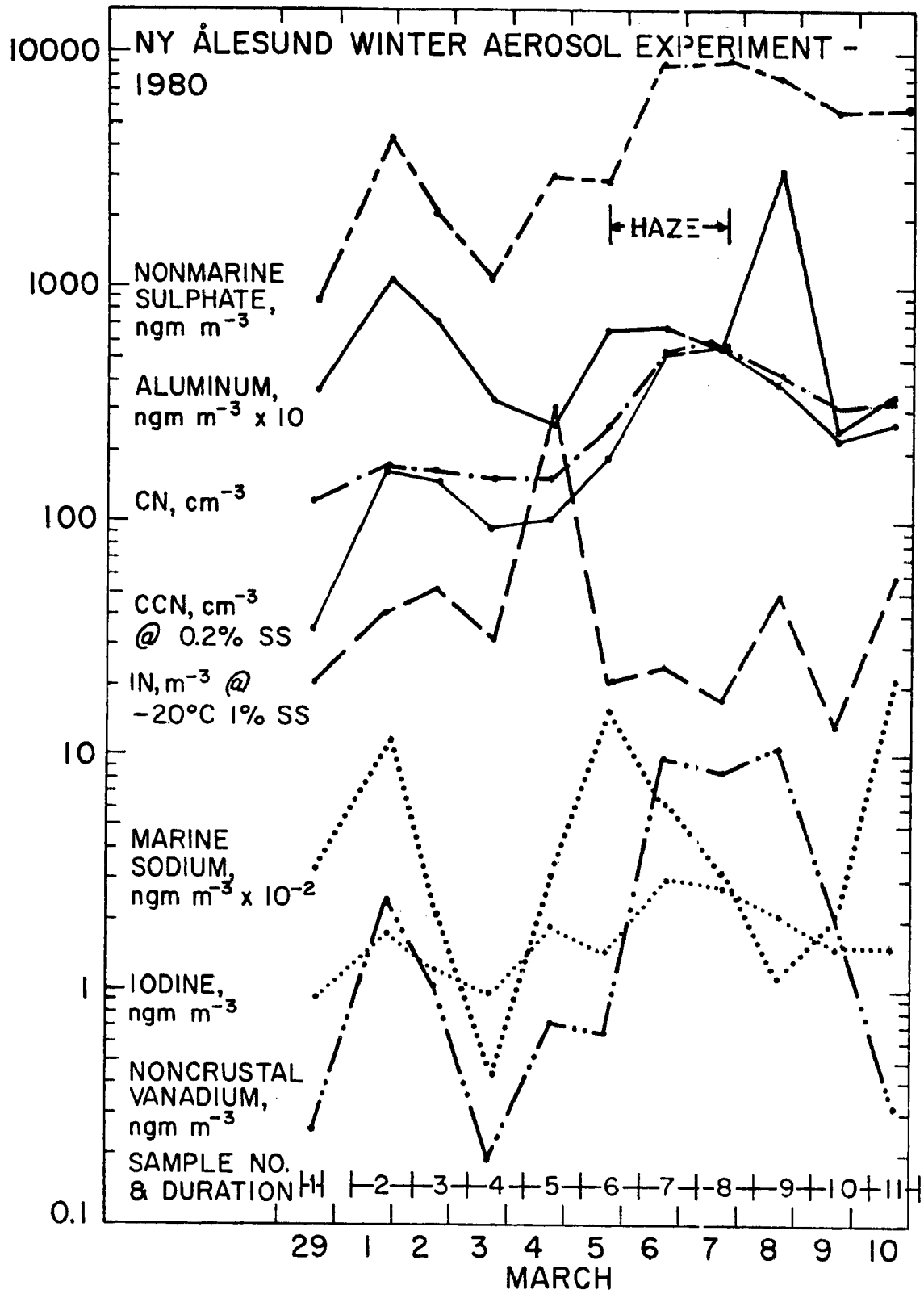


Fig. J.5 Ny Alesund, Spitsbergen, Norway winter aerosol experiment, February - March 1980.

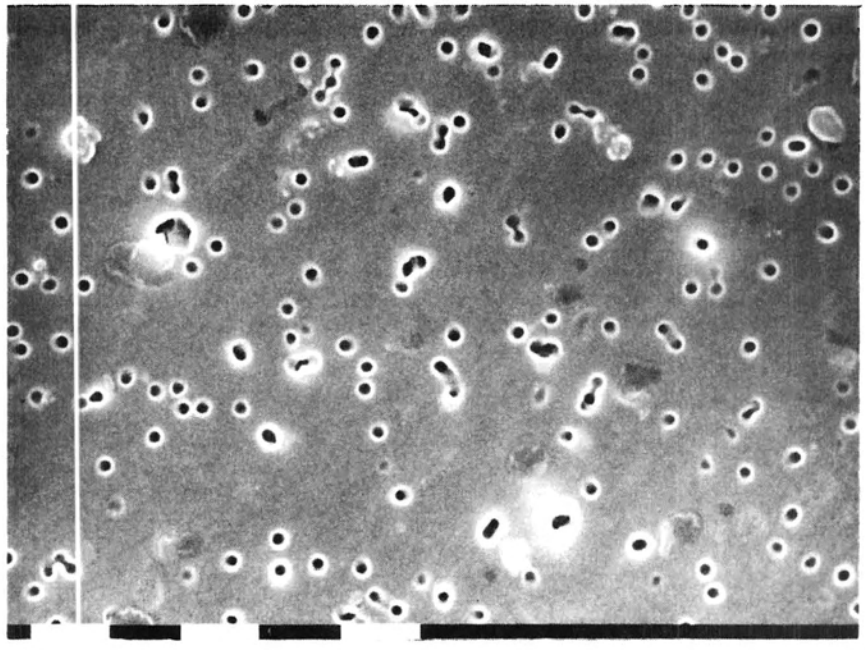


Fig. J.6a Scanning electron microscope photographs of aerosol particles collected at Ny Ålesund during winter on 0.2 μm diameter pore size Nuclepore filters from sample #4, 3100X magnification, bar scale is 10 μm.

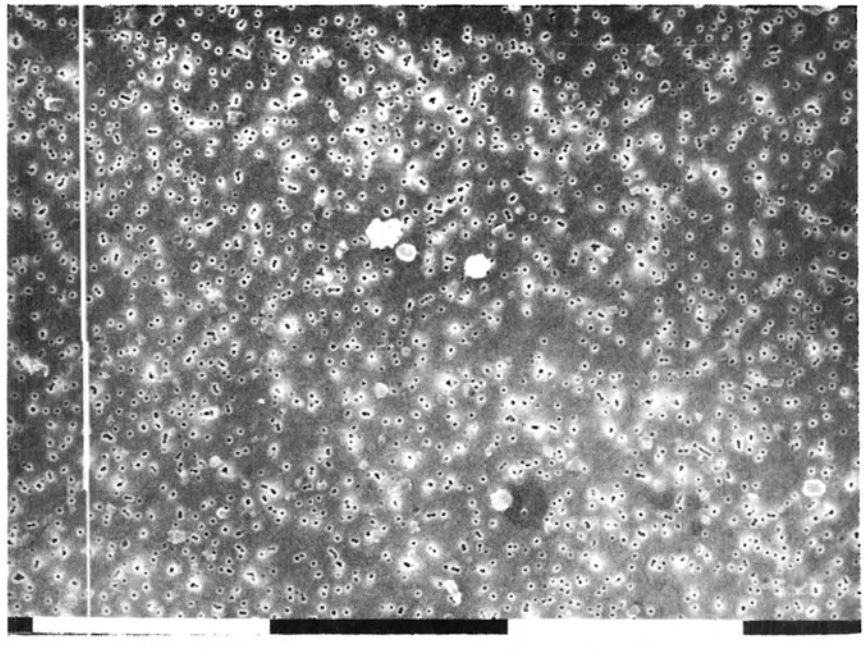


Fig. J.6b Same as a but 10,000X magnification, bar scale is 1 μm.

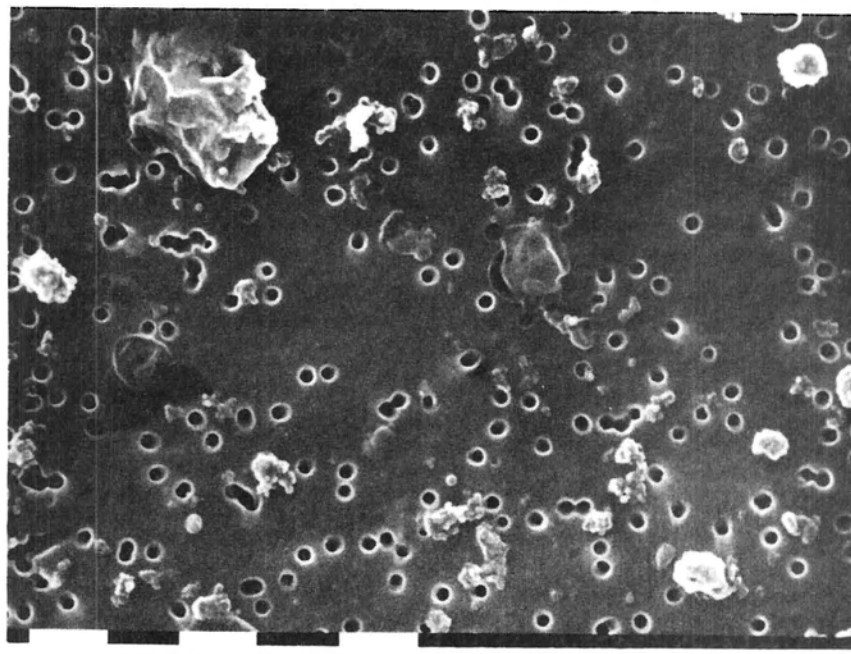


Fig. J.6c Same as a, but for sample #8.

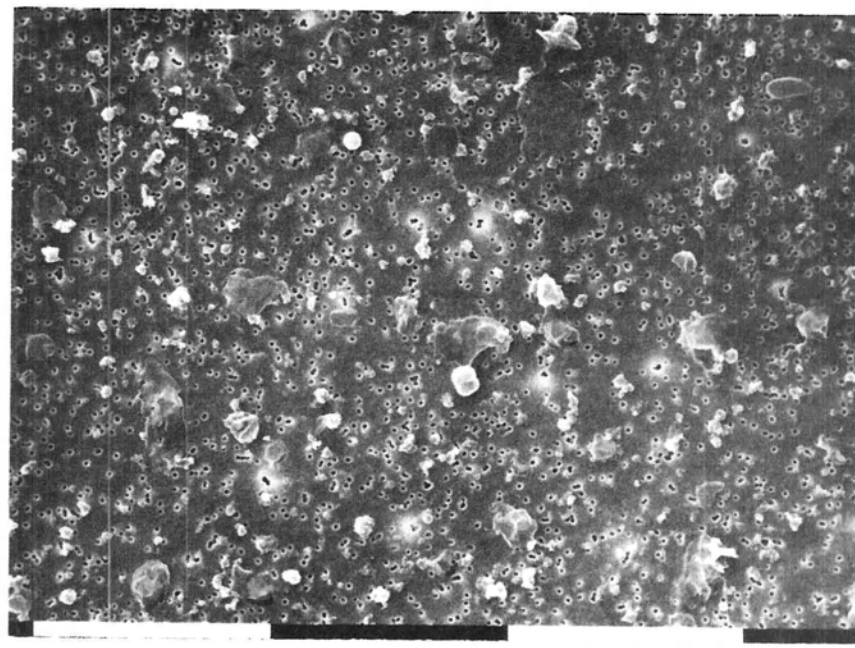


Fig. J.6d Same as c, but 10,000X magnification, bar scale is 1 μm.

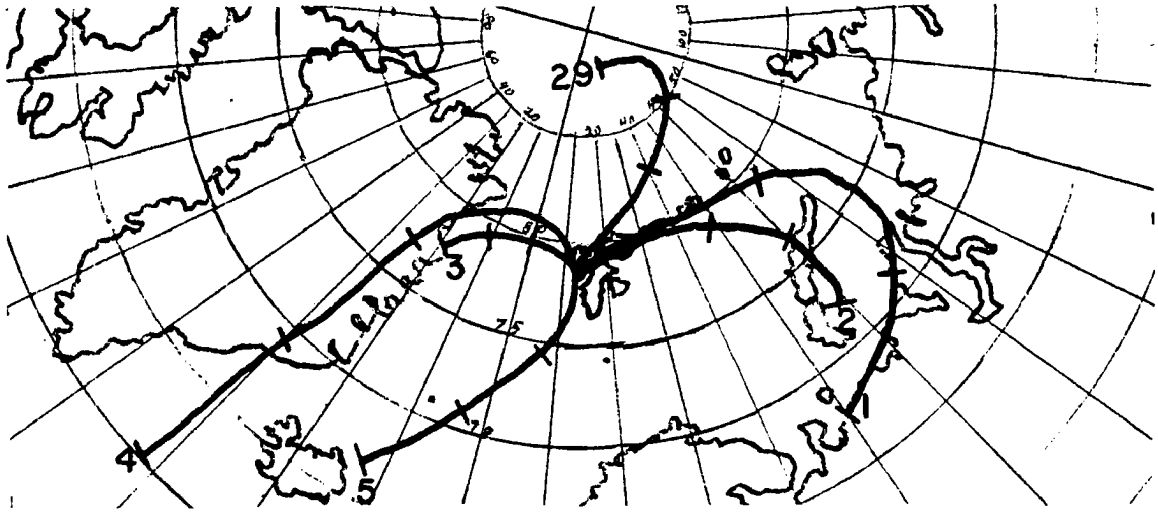


Fig. J.7b Same as a at 700 mb.

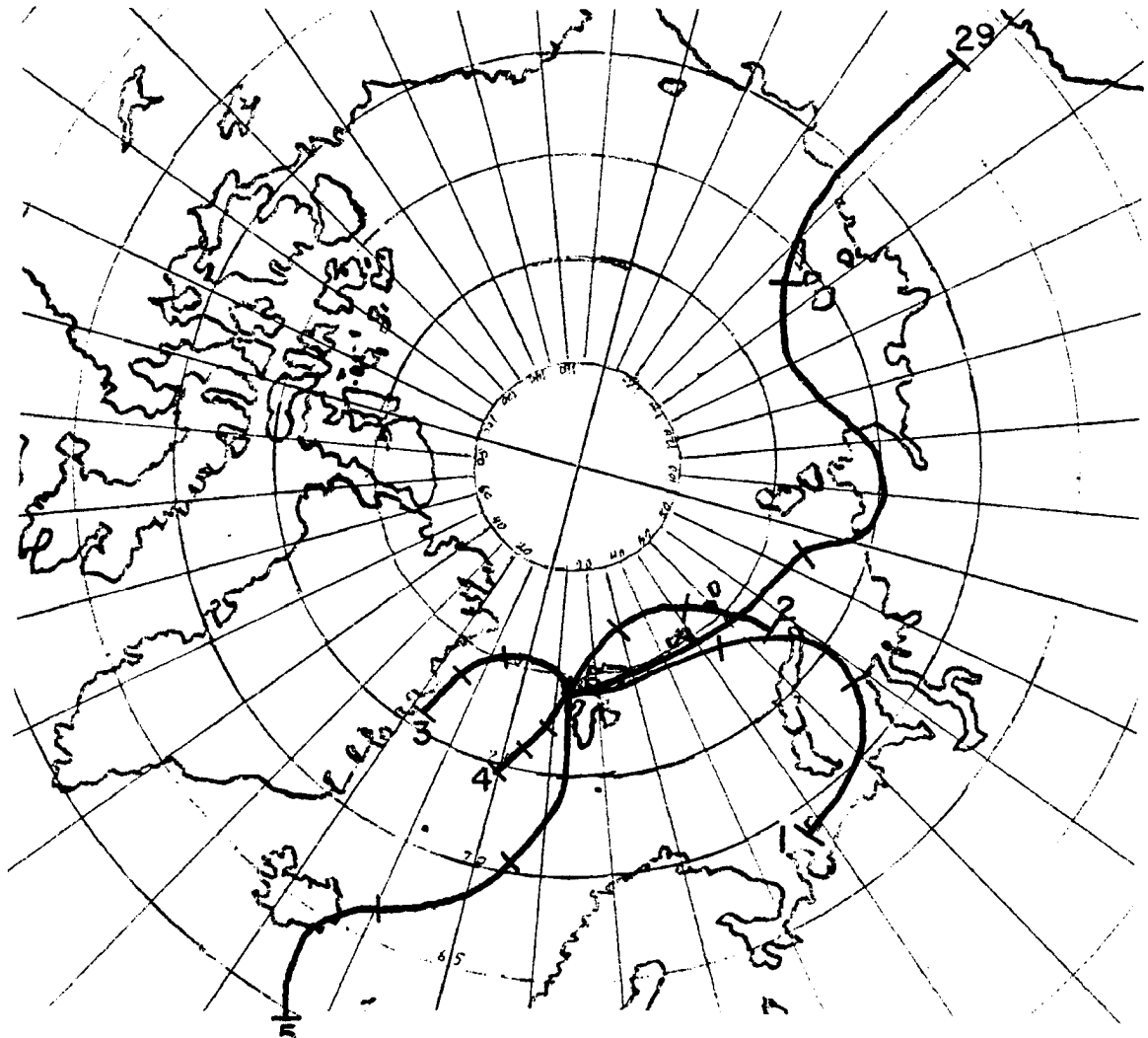


Fig. J.7a Ny Ålesund 850 mb isohypse analyses, 29 February – 5 March, 1980, valid at 0Z on date indicated on each isohypse. Each tick mark represents 24 hr. movement upwind based upon winds at the start.

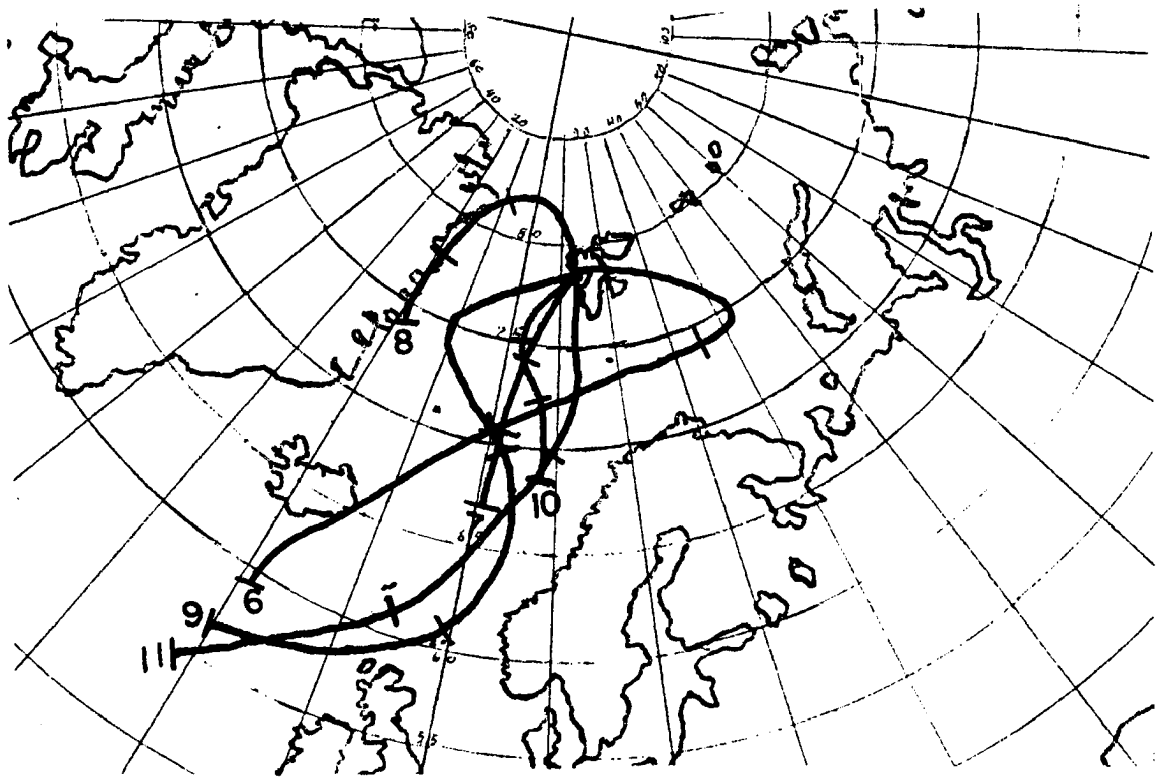


Fig. J.7d Same as a at 700 mb, 6 - 11 March, 1980.

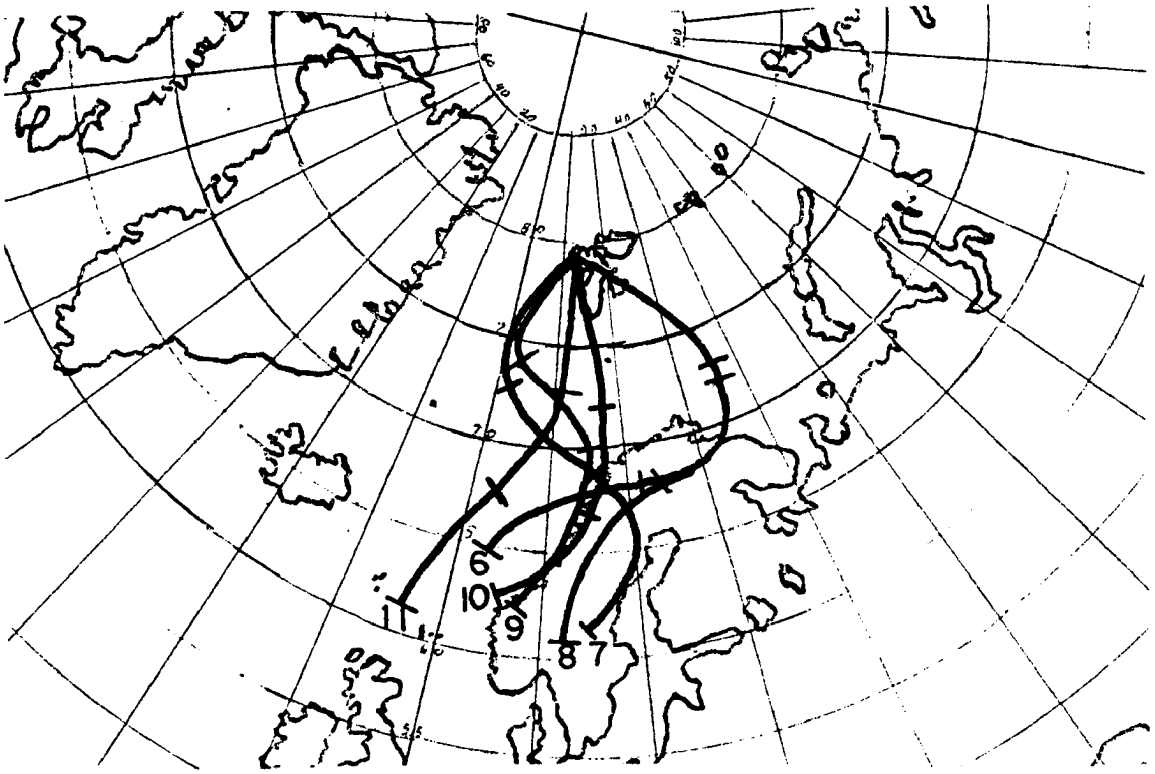


Fig. J.7c Same as a at 850 mb, 6 - 11 March, 1980.

and 8. Figures J.7a-d are the isohypse analyses for the 850 mb and 700 mb pressure levels for the period of the experiment.

The prevailing wind direction at the surface was from the southeast during the experiment. The winds aloft as seen in Figure J.7 were variable and occurred from all directions during the study period. This is similar to the conditions experienced at Barrow during the winter where surface wind conditions were also not representative of the overall synoptic situation. The site is within the arctic polar boundary layer and under the influence of the polar easterlies at the surface. The change in wind direction aloft is recognizeable at the surface in the change in the composition of the aerosol.

Although Ny Alesund is located at 79° N, the ocean around the site was ice free. The warm Gulfstream waters maintain this region ice free throughout the winter. There is, as a result, the presence of fresh marine aerosol particles at all times, increasing in concentration with wind speed. This is the cause of the three peaks in marine Na in samples 2, 6 and 11 as seen in Figure J.5.

The results of the aerosol chemistry analyses plotted in Figure J.5 can be divided into three sections. The concentrations of the pollutant aerosol tracers, $SO_4^{=}$ and V, as well as Al and CN go through a maxima at the beginning and end of the period with minima occurring on sample 4. This can be explained in terms of the direction from which airmasses are being advected to the site. Figure J.7 shows the winds were east to north during the first tracer maximum. This was the coldest period with temperatures ranging from -20°C to -25°C. This period is representative of a true arctic airmass during the winter. Concentrations of V are about three times higher than those measured at Barrow during the

winter. Non-marine $SO_4^=$ has about the same concentration as Barrow during the winter. The pollution character of the arctic aerosol is evident at this time.

The concentration of these pollutant indicators then drops to a minimum on sample 4. The isohypse analyses indicate the winds changed to west to southwest, a direction from which an airmass more representative of the North Atlantic is advected to the site. The aerosol-particles collected at this time can be seen in Figures J.6a and b. The study site at Irafoss, Iceland is upwind at this time. The concentrations of the aerosol do not reach the extremely low values measured at Irafoss suggesting there is a mixture of aerosol sources represented during these samples. This is the " clean air" direction for this site during the winter.

The winds continue to back with time and turn to south to southeast. By sample 7, a polluted airmass is obviously present. Restricted visibility due to haze was reported. The pollutant aerosol tracers V and $SO_4^=$ reach their maximum, factors of 10 and 2.5 times higher, respectively, than those measured at Barrow during winter. This is representative of transport directly from the pollution aerosol source region of Europe and the western Soviet Union. This period shows the magnitude of pollutant aerosol transport to the north at this location. These high concentrations of pollutants are present after a transport distance of approximately 3000 km. Ny Alesund is located 1200 km from the North Pole. It would not be difficult to imagine the further advection of this kind of pollutant aerosol from Europe well into the Arctic Ocean basin. Figures J.6c and d show the aerosol particles collected during sample 8. Fly ash particles can be seen, as

well as the low contrast particles which are probably primarily composed of the light elements (at wt. <11) and secondary or organic in nature.

J.2a Cloud condensation nuclei

As expected from the results of other arctic locations where there is a strong anthropogenic signal in the aerosol composition, the CCN are highly correlated with the pollutant aerosol indicators noncrustal V and nonmarine $SO_4^=$. Table 5.2 shows the CCN are most strongly associated with the secondary pollutant, $SO_4^=$, a result of gas to particle conversion along the transport path of SO_2 emissions. The high correlation with CN is a result of the dominance of the total aerosol by the hygroscopic pollutants. The CCN at 0.2% SS in the mean at this site accounted for 83% of the total aerosol particle numbers. During the maximum pollutant aerosol concentrations, the CCN at this low supersaturation could account for 95% of the total aerosol number. This is indicative of the highly secondary nature of the aerosol.

Also evident from Table 5.2 is the dependence of the CCN on the secondary marine aerosol tracer, I. This was also true at Irafoss, 2000 km to the southwest of Ny Alesund, which is located in essentially the same marine environment of the North Atlantic and under the influence of the Gulfstream. This was observed in Narrangansett and to a lesser extent at Poker Flat. The true nature of this relationship is not known, but warrants further investigation into the importance of possible secondary aerosol particles from marine sources on the maritime concentrations of CCN. Finally, although marine Na was present, there is no indication that primary marine aerosol or sea-salt particles are an important component of the total CCN concentration.

The CCN concentration during the first maximum resulting from advection from the north are approximately the same as those observed in Barrow during winter. With flow from the south, the CCN concentration increases by a factor of three. However, advection from both directions brings the site under the influence of anthropogenic aerosol. Ny Alesund is the most polluted of the arctic study sites.

J.2b Ice nuclei

The IN vary over an order of magnitude during the study period. Maximum concentrations are about the same as observed during the YMER-80 study when under the influence of anthropogenic aerosol from the south. The similarity ends there. The IN concentrations do not show any relationship to the anthropogenic aerosol particles present during the winter. Table 5.2 shows the IN do not covary with any of the aerosol elemental source tracers. The isolated peak of sample 5 cannot be attributed to any unusual event during the sampling. The lack of any relationship to any source tracers in the presence of such strong signals by the measured tracers suggests the competing effects of more aerosol numbers and the possible 'poisoning' of active IN by pollutants. The result is an IN population that has a random day-to-day variation that cannot be attributed to any measured aerosol or meteorological transport factor.

J.3 Seasonal Differences in Cloud Active Aerosol

Figure J.8 presents the seasonal mean spectra for CCN for the experiments conducted on board the YMER during summer and at Figures J.9 and J.10 depict individual IN concentrations for summer and winter, respectively, with the mean spectra plotted as solid lines. Ny Alesund during winter. Again there was difficulty with the IN determinations at

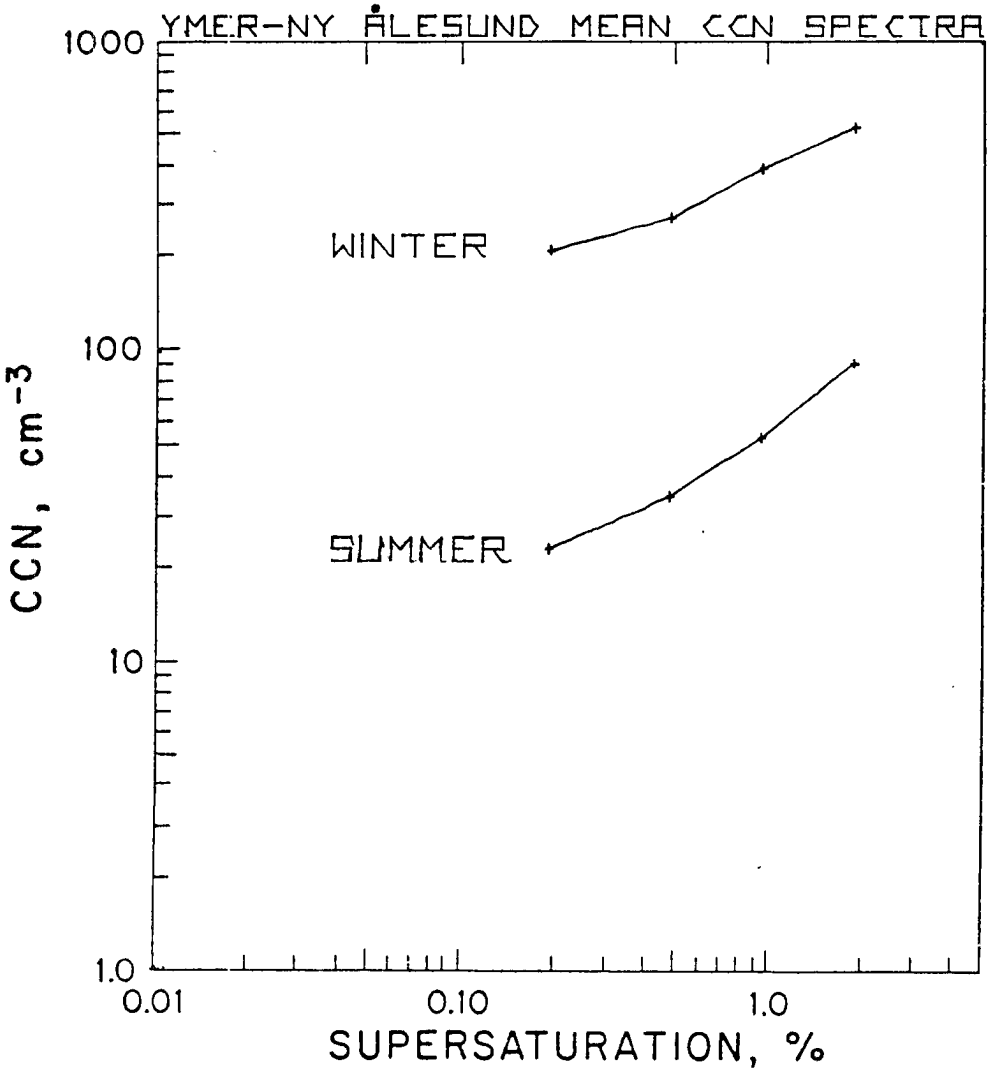


Fig. J.8 Mean CCN spectra for the YMER-80 cruise derived from thirty-one spectra measured during the summer, 11 August - 1 September, 1980 and for Ny Alesund derived from twenty-two spectra measured during winter, 28 February - 11 March, 1980.

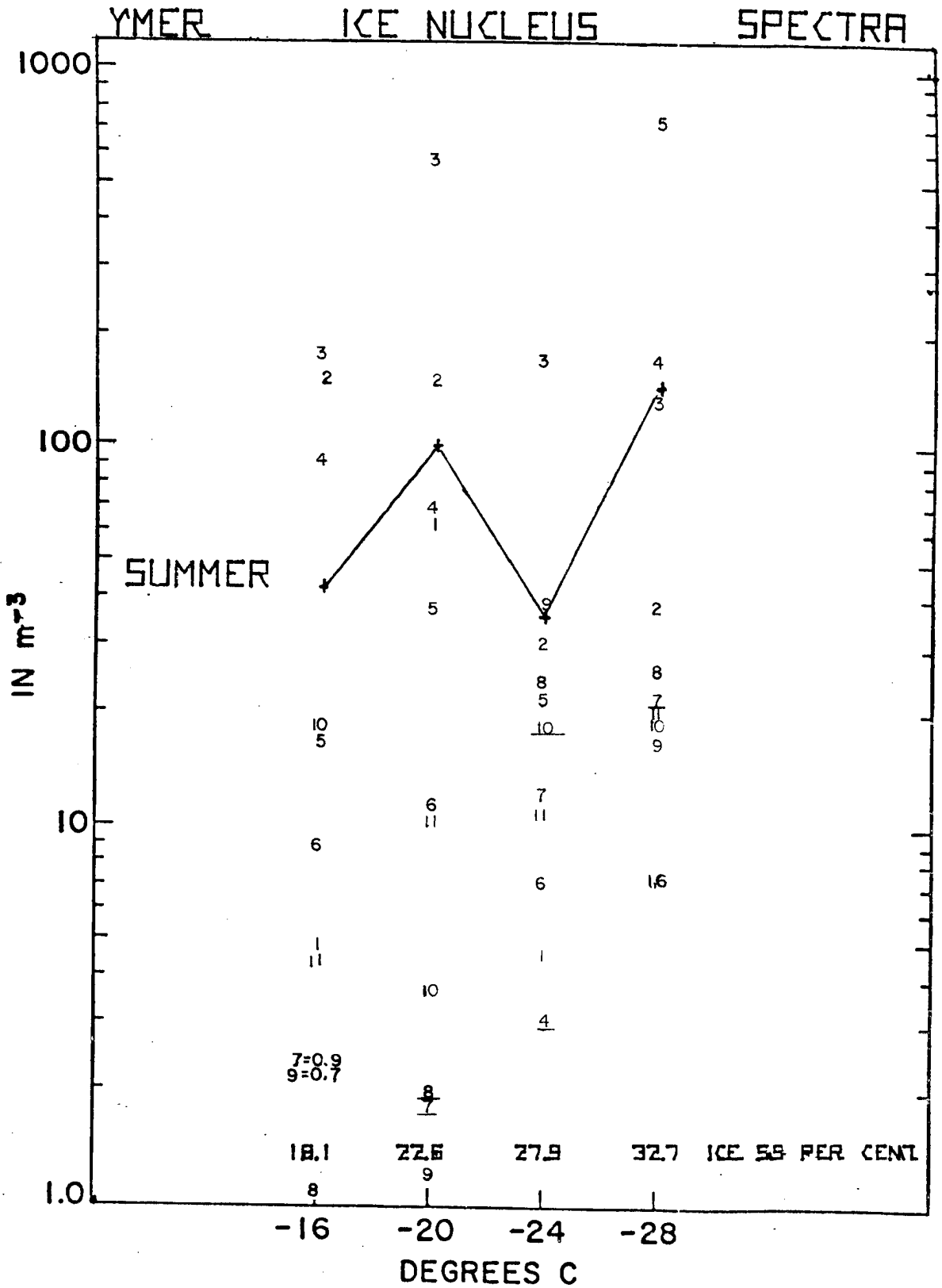


Fig. J.9 IN spectra for the YMER-80 expedition, summer experiment, 11 August - 1 September 1980. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank concentrations. The mean spectrum is plotted using only values above blank.

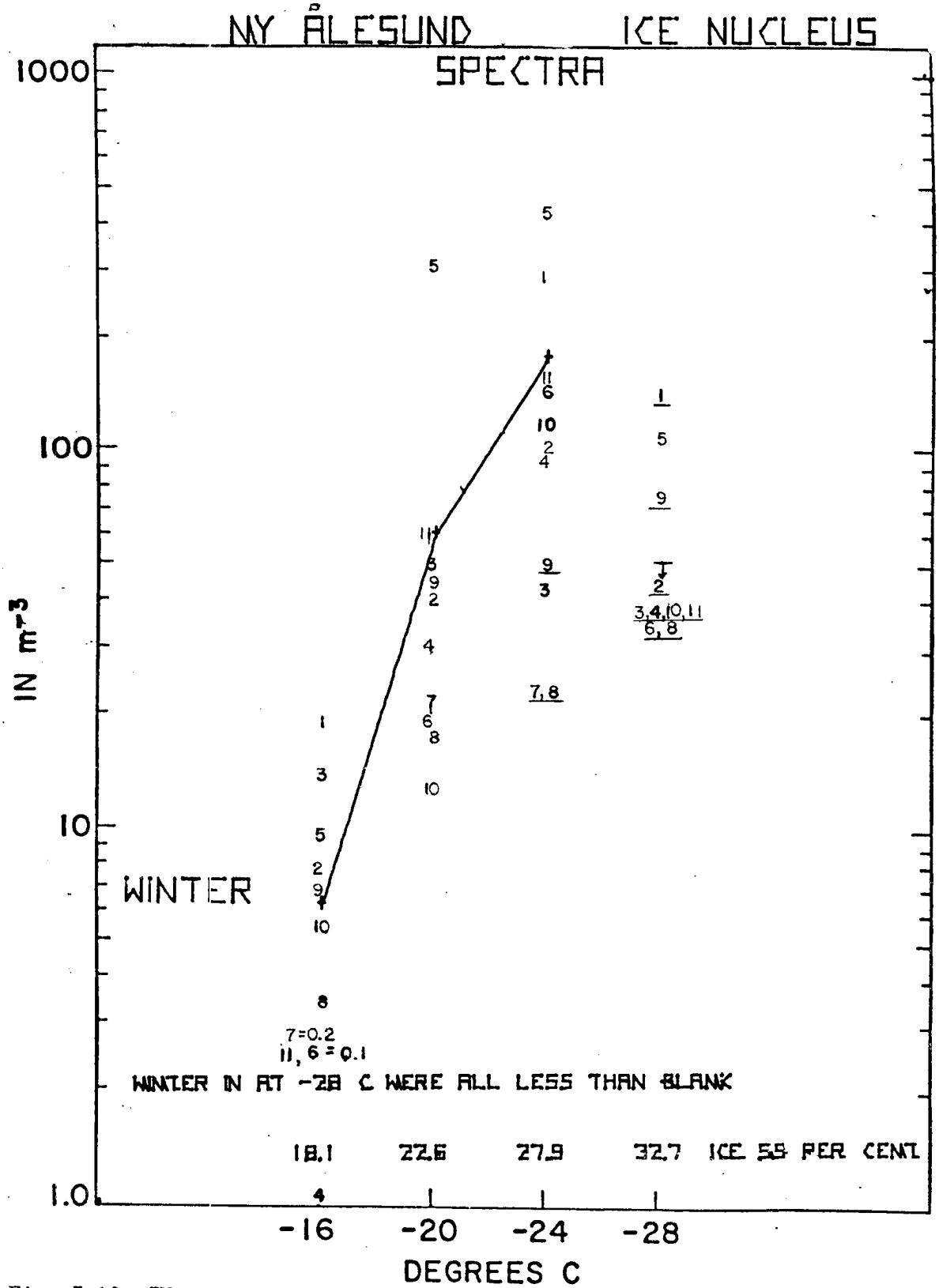


Fig. J.10 IN spectra for NY Alesund, Spitsbergen for the winter experiment, 28 February - 11 March 1980. Concentrations at each temperature for each sample are indicated by the sample number. Underlined samples indicate less-than-blank

$-28^{o}C$ due to high blank filter values. The summer IN spectra had a great deal of scatter as evidenced by the seasonal mean spectra. The two best determinations at $-16^{o}C$ and $-20^{o}C$ show the winter time values to be depressed in comparisn to the summer values. Referring to Table 5.4 the winter to summer ratios of the pollutant aerosol tracers are compared to the IN seasonal ratios. The Ny Alesund results are similar to the results from Barrow and Narragansett. There is an inverse relationship between the IN and the pollutant aerosol average seasonal concentrations. Figure J.9 and J.10 show the effect is most pronounced at the lower ice supersaturations. This was also the case at the other North Atlantic site of Irafoss, Iceland. This suggests the IN active at warmer temperatures are more susceptible to the possible 'poisoning' effects of air pollutants, or are more likely to be preferentially removed as a result of colder temperatures during winter. Temperatures during winter at Ny Alesund averaged $-16^{o}C$, $20^{o}C$ colder than the average temperature of the YMER-80 expedition (see Table 5.1).

The CCN at Ny Alesund-YMER show the greatest seasonal difference in their mean spectra of all the arctic sites. This is comparable to the seasonal difference of the pollutant aerosol tracers as shown in Table 5.3. Again, the largest difference is in the lower supersaturation range, producing a flatter spectra indicating the presence of higher concentrations of haze forming nuclei during the winter. Haze was observed during the winter experiment for several days. The strong seasonal dependence of pollutant concentrations is reflected in the seasonal difference in the CCN at this site.

J.4 Sources of Cloud Active Aerosol

From the previous discussion it is readily apparent that the European source region is of primary importance at this study location as a source of CCN. It is a source of CCN both during the summer and winter. Stronger circulation during winter results in a more significant flux of CCN, or their precursors, at this time. This is not the case for IN. There are certainly sources of IN within the source region of Europe and Eurasia. It is apparent that interaction within this polluted atmosphere of particles and gases serves to mask their effectiveness. In terms of long range transport, the region as a whole does not act as an IN source during winter. This may be a result of pollutant 'poisoning', or preferential removal due to colder winter temperatures. Results from the summer experiment show that the transport of aerosol particles from the continent to the north acts as a relative source of IN because of the very clean background conditions present in the Arctic during summer. In spite of this, summer concentrations of IN are higher in the mean.

The most important result of the study was the elimination of transport of cloud-active aerosol and pollutants from North America across the North Atlantic Ocean as a possible source to the Arctic. This was expected, based upon the similar results from the studies at Irafoss, Iceland. The Eurasian source is the most important source of pollutants for the Arctic.

There is also evidence of an important secondary marine component of the CCN advected to the Arctic at this location. What fraction of the total may be ascribed to this source is difficult to assess. No marine component of IN was found, unlike the results from Barrow. The

difference in the biological activity in the ocean waters surrounding these sites may be a reason for these differences. This can only be speculated upon at this time.